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STATUS REPORT NO. 5 ON
CLINCH RIVER STUDY

Editor
R. J. Morton

NOTICE

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HEALTH PHYSICS DIVISION

STATUS REPORT NO. 5 ON CLINCH RIVER STUDY
Clinch River Study Steering Committee

EDITOR

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OCTOBER 1965

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

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**Succeeded Vincent Schultz as an ex officio member of the Steering Committee effective August 20, 1964.

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*Succeeded A. G. Friend (see footnote, page vii).
**Succeeded P. H. Carrigan, Jr. (USGS).
***Succeeded D. B. Porcella (USPHS).

ACKNOWLEDGMENT

As explained for studies described in previous status reports on the Clinch River Study, essential parts of the work are performed by a number of groups in the Oak Ridge National Laboratory and other agencies represented on the Steering Committee. The Committee recognizes and appreciates participation of the agencies and individuals named below.

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and

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S. Leary Jones

For the TENNESSEE GAME AND FISH COMMISSION:

L. P. Wilkins

FEATURES OF THE OVER-ALL PROGRAM

The four status reports previously issued have been based primarily on progress reports submitted at meetings of the Clinch River Study Steering Committee.[1,2,3,4] This fifth status report is mainly a summary of the study from April 26, 1962 to December 4, 1963. During this period the Steering Committee met twice, in February and December 1963, and received the progress reports upon which this report is largely based.

The cooperative nature and the basic objectives of the Clinch River Study were restated in the beginning of Status Report No. 4.[4] The agencies and individuals who have contributed to the study during the period of the present report are indicated in the acknowledgment on pages xi-xiii, and the list of contributing authors is on the title page.

Steering Committee Actions

The Steering Committee held meetings on February 6 and 7, 1963 at the R. A. Taft Sanitary Engineering Center of the U. S. Public Health Service (USPHS) in Cincinnati, Ohio. These included two open sessions for progress reports and discussions, a tour of the USPHS laboratories where radiological analyses of water samples from the Clinch River Study were made, and an executive meeting of the Steering Committee in the afternoon of February 7.

The meetings in December 1963 were at the Oak Ridge National Laboratory (ORNL). They included two open sessions and a demonstration of the counting equipment used in analyses of bottom-sediment core samples on December 4, and an all-day executive meeting December 5, 1963.

Open Meetings

February 6-7, 1963 (Cincinnati, USPHS). — Two open sessions in the mornings of February 6 and 7 were attended by a total of 37 persons. Eight summaries on specific work projects and progress reports by the four subcommittees of the Steering Committee were presented and discussed as follows:[5] (1) "Report of Applied Health Physics Surveys in 1962," by H. H. Abee (ORNL); (2) "Progress Report No. 3, Subcommittee on Water Sampling and Analysis," by M. A. Churchill (TVA), Chairman; (3) "Hydrologic Measurements and Analyses," by R. M. Richardson, P. H. Carrigan, Jr., J. P. Monis, and R. J. Pickering (all of USGS); (4) "Data Evaluation," Progress Report of Subcommittee on Bottom Sediment Sampling and Analysis, by P. H. Carrigan, Jr. (USGS), Chairman;* (5) "Statistical Analysis of Sediment Samples Collected in June, 1961," by K. G. Busch (USPHS); (6) "Data on Sediments Collected in June and August 1961," by R. W. Andrew (USPHS); (7) "Progress Report on 1962 Bottom Sediment Coring Program," by R. J. Pickering (USGS) and P. H. Carrigan, Jr. (USGS) ; (8) "Use of the Swedish Foil Sampler for Taking Undisturbed Cores of River Bottom Sediments," by R. J. Pickering (USGS); (9) "Progress Report No. 2, Subcommittee on Aquatic Biology," by S. I. Auerbach (ORNL), Chairman; (10) "Data on Fish Collected in June and December 1961 and in March 1962," by D. B. Porcella (USPHS); (11) "A Pilot Study to Determine the Distribution in Space and Time of Multiple

*R. M. Richardson (USGS) later succeeded P. H. Carrigan, Jr. as subcommittee chairman. (See subcommittee membership on page x of this report.)

Radionuclides in a Seminatural Pond Environment," by William A. Brungs, Jr. (USPHS); and (12) "Progress Report No. 2, Subcommittee on Safety Evaluation," presented by C. P. McCammon (TDPH), Chairman. The report on safety evaluation studies was covered by three speakers introduced by McCammon as follows: "Application of MPC_w to ORNL-Discharged Radionuclides in Clinch River," by W. S. Snyder (ORNL), "Preliminary Evaluation of ORNL Waste Water Discharges to Clinch River," by K. E. Cowser (ORNL), and "Whole-Body Counting of ORGDP Employees," by F. L. Parker (ORNL).

December 4, 1963 (ORNL). — The open meeting was attended by a total of 27 persons. The programs of the four subcommittees were covered by progress reports as outlined below:[6]

A. Water Sampling and Analysis. - M. A. Churchill, Chairman

"Dye Tracer Studies - Summer 1963" by B. J. Frederick (USGS), F. L. Parker (ORNL), and P. H. Carrigan, Jr. (USGS); "Status of Analyses of Water Data," by R. W. Andrew (USPHS) "Stable Chemical Composition of the Clinch and Tennessee Rivers," by R. J. Pickering (USGS); and "Extraneous Sources of Release of Radionuclides to Clinch River," by R. J. Pickering, P. H. Carrigan, Jr., and W. M. McMaster (all of USGS).

B. Bottom Sediment Sampling and Analysis. - R. M. Richardson (USGS), Chairman, presented by P. H. Carrigan, Jr. (USGS).

"Bottom Sediment Cores Collected during the Summer of 1962," by R. J. Pickering (USGS); "Behavior of Radionuclides Associated with Clinch River Sediments," by W. P. Bonner (ORNL) and T. Tamura (ORNL); "Relationships in the Longitudinal Distribution of Radionuclides in the Clinch and Tennessee Rivers," by P. H. Carrigan, Jr.

(USGS); "Seasonal Changes in the Longitudinal Distribution of Radioactivity in the Bottom Sediments of the Clinch River," by P. H. Carrigan, Jr. (USGS); "Radiation Levels in Sloughs," by R. J. Pickering (USGS); "1963 Applied Health Physics Survey," by H. H. Abee (ORNL); and "Clinch River Channel Improvements: A Study of Radiological Hazards," by O. W. Kochtitzky (TVA) and H. H. Abee (ORNL).

C. Aquatic Biology. - S. I. Auerbach (ORNL), Chairman

"Use of Specific Activities in Predicting the Uptake of ^{90}Sr by White Crappie (*Pomoxis annularis*)" by D. J. Nelson (ORNL); and "Movement of Smallmouth Buffalo (*Ictiobus bubalus*) in the Vicinity of White Oak Creek," by D. J. Nelson (ORNL).

D. Safety Evaluation. - C. P. McCammon (TDPH), Chairman

"Status Report: Estimates of Radiation Exposure Doses to Man from Radionuclides Released to the Clinch River," by K. E. Cowser (ORNL).

Summation. - A summary of the information covered during the open meeting was presented by F. L. Parker (ORNL), Study Coordinator. For this summation and all the previous reports time was allowed for discussion and questions concerning the information presented.

Executive Meetings

February 7, 1963 (Cincinnati, USPHS).[7] - At this meeting the Steering Committee approved release of information and data in a paper "Use of the Swedish Foil Sampler for Taking Undisturbed Cores of River Bottom Sediments," by R. J. Pickering (USGS), for presentation at the Federal Interagency Sedimentation Conference, Jackson, Mississippi, January 28-February 1, 1963, and for publication in the proceedings of the Conference by the U. S. Department of Agriculture.

Most of the time at this meeting was devoted to program plans and recommendations presented by the chairmen of the four subcommittees. These are mentioned briefly below and given in more detail in the minutes of the meeting.[7]

Water Sampling and Analysis. - M. A. Churchill (TVA), Chairman. The chairman referred to pages 51 and 52 in the subcommittee's Progress Report No. 3, presented the day before.[8] Six recommendations were outlined and explained: (1) proportional sampling of Clinch River at Centers Ferry; (2) intensive surveys at several stations on Clinch River after Melton Hill Dam is in operation; (3) study of findings at the Centers Ferry station and decision whether there is need for additional stations; (4) establishment of a network of stations, if necessary, for monitoring radioactivity in precipitation (rain or snow) in connection with the water sampling program; (5) resolution of the problem of disagreements in analytical results between different laboratories presumably using comparable procedures on duplicate water samples; and (6) no analyses of river water samples for stable chemicals in any future water sampling studies.

Bottom Sediment Sampling and Analysis. - P. H. Carrigan, Jr. (USGS), Chairman. The subcommittee recommended and the Steering Committee approved the appointment of James W. Beverage (TVA) as a member of the subcommittee to succeed James Smallshaw of TVA who had retired. Also, H. H. Abee of the ORNL Applied Health Physics Section was made a full member of the subcommittee. (See page x).

Recommendations concerning work plans and investigations included the following: (1) joint work by the subcommittee and the Applied Health Physics Section to develop recommendations for future monitoring of bottom sediments in the river; (2) several studies (which were described) to provide information on retention of specific radionuclides by the various types of sediments in suspension or after deposition;

(3) extensive geochemical studies to delineate the physicochemical processes that lead to incorporation of radionuclides in bottom sediments; (4) cooperation of this subcommittee with other groups or subcommittees in studies of the physicochemical state of radionuclides in the bottom sediments and also in the water of White Oak Creek and Clinch River (including suspended sediments); (5) continued efforts to better define the phenomena of desorption and migration of radionuclides in sediments and of equilibrium conditions in the river; and (6) closer alliance of this subcommittee's work with that of the Subcommittee on Aquatic Biology.

It was the consensus of the Steering Committee that the proposed plan was too ambitious for the time remaining to complete the Clinch River Study but it was agreed that in general the program should be developed as recommended.

Aquatic Biology. - S. I. Auerbach (ORNL), Chairman. The subcommittee recommended: (1) Collect another series of carpsucker and buffalo fish from the Clinch River, primarily to obtain data from different methods of sample processing. Several variations of sample processing were suggested. (2) Acquire information on the quantity, kind, method of preparation, and rate of consumption of Clinch River fish eaten by people living nearby and fishing almost exclusively in the Clinch River. (3) Obtain estimates of the per capita consumption of fish in Tennessee and also, as specific information as possible regarding the kinds and quantities of fish sold in the East Tennessee areas and in Nashville and Memphis. It was recognized that to obtain accurate data regarding fish consumption is difficult.

A representative of the subcommittee on Safety Evaluation explained that for the safety evaluation maximum individual values are desired both as to consumption of fish and concentrations of radionuclides. If well-verified average values can be obtained the maximum is estimated as greater than the average by a factor of 3.

It was noted that radioactivity in fish from contaminated waters in a particular area would be diluted during marketing and consumption because fish from uncontaminated areas would be bought and mixed with them.

Safety Evaluation. - C. P. McCammon (TDPH), Chairman. The recommendations of this subcommittee were mainly about needs for data from actual measurements which would make the estimates of potential radiation exposures from various media more reliable. In brief, the subcommittee suggested that work be done to provide: data from laboratory analyses in progress (on water, sediments, and fish); measurements relative to radionuclides involved in crop irrigation; measurements relative to water immersion and contact with contaminated sediments; and the potentially exposed population from estimated water usage.

Questions were raised as to the need to obtain various data regarding levels of radiation, for example, from sediments in embayments, radionuclides accumulated in food through irrigation, and recreational use of Tennessee River reservoirs. It was agreed that under present conditions there is no significant hazard from any of these potential exposures but that methods for evaluating them should be developed as possible so that future changes can be studied more reliably. A number of gaps in the information presently available were mentioned and discussed.

The Steering Committee chairman commented that data needed by the Safety Evaluation group should be provided in so far as practicable. He suggested that attempts be made to obtain information about: (1) metabolism differences,

(2) techniques for the interpretation of data on soil and food in relation to irrigation hazards, (3) a restudy of fish using the analytical techniques already worked out but with different methods of processing the fish samples, (4) data from field measurements of radiation emitted by bottom sediments, (5) measurements of radiation in water sufficient to indicate the immersion dose, and (6) further study of assumptions and methods for calculating dose rates from various exposure media. It was agreed that during the next several months of the study efforts would be made to obtain data on these conditions.

Other Discussions and Actions. - For the information of the Steering Committee and as a matter of record Chairman E. G. Struxness discussed changes initiated at ORNL to improve the system of control and minimize discharges of radioactive wastes. He reviewed a memorandum from the ORNL Waste Effluents Committee to management officials of the Laboratory on February 1, 1963 which outlined activities and major recommendations of that committee during the previous year.

Radioactivity released to the river during 1962 was less than in 1961 and further reduction was expected. Nine "major recommendations" for control facilities and for operational and communication procedures were outlined. These included an evaporator for intermediate-level liquid wastes which had been authorized and was under construction, plans for backfilling the intermediate-level waste pits and covering them to prevent continued seepage, and construction plans for a dam and other works, if necessary, to control the outflow from White Oak Creek to Clinch River after Melton Hill Dam is in operation. There were several specific recommendations for improvement of intercommunication among groups in the Laboratory and, concurrently, the development of safety evaluations and statements of permissible limits of discharges of radionuclides so that the advice of the ORNL health physicists might be utilized more effectively.

The Steering Committee discussed the time for completion of investigations and a final report on the Clinch River Study. It was the consensus that the study must include conditions in so-called "Phase II" (after Melton Hill Dam is in operation), that the remainder of the study should be covered by status reports including Phase I and whatever investigations were made in Phase II, and that a final report on the whole study would be necessary. The preparation and issue of Status Report No. 4 was authorized.

The problems of controlling the use by others of preliminary data from the study were discussed thoroughly. It was decided that informal progress reports prepared for the Steering Committee should be marked clearly to restrict publication or quotation without express permission from the authors and approval by the Steering Committee. It was agreed that independent analyses of the data presented in progress reports should be made only under the auspices of the Steering Committee and as a part of the Clinch River Study program. It was also agreed that status reports published in the future should be restricted by a printed notice that the information was of a preliminary nature and should not be republished without written permission.[7]

Meeting December 5, 1963 (ORNL). - The one-day executive meeting on December 5 was devoted mainly to reports and recommendations from the four subcommittees. These reports, summarized briefly below, are given in more detail in the minutes of the executive meeting.[9]

A. Water Sampling and Analysis. - The subcommittee chairman, M. A. Churchill, submitted the following recommendations:

1. The TVA should be asked to shut off flow at Melton Hill Dam completely for as long as possible, say a week or more, and the increased concentrations of radioactivity in the river expected to result from complete shutdown of flow at the dam should be studied.

2. For future monitoring of the Clinch River at least one continuous water monitoring station downstream from White Oak Creek should be established and maintained. It was stated that a station at about CRM 14.5 (near ORGDP) would be preferable; and that the cost at that location had been estimated as not more than $5,000 and probably less.

3. Based on indications that Melton Hill Dam will not cause much change in the Tennessee River at Watts Bar Dam or below, the basic water sampling program should not be reestablished as part of Phase II of the Clinch River Study.

4. When revision of the radiological data (then in progress) has been completed and the subcommittee progress report rewritten, the data on water contamination will be sufficient to complete Phase I of the Clinch River Study.

5. Every possible effort should be made to operate Melton Hill Dam so as to simulate winter releases, and to extend the program of tracer tests in order to investigate dispersion in the river under winter conditions.

The Steering Committee agreed that these five recommendations should be accepted and followed in so far as practicable.

B. Bottom Sediment Sampling and Analysis. – Subcommittee chairman, R. M. Richardson was not present and the report was presented to the Steering Committee by P. H. Carrigan, Jr. Several reports that were being prepared to complete Phase I were mentioned. Also the subcommittee outlined the studies proposed for Phase II of the Clinch River Study. These included:

1. Sediment surveys to represent the beginning of Phase II and later periodic sediment surveys, to be made by the TVA Hydraulic Data Branch.

2. Studies to identify trends in the movement of bottom sediments by special techniques with various marker materials, for example, the use of fluorescent calcite deposited on the bottom and identified fluorometrically in sediment samples collected later to investigate scouring or relocation of sediments.

3. Continued geochemical studies of bottom sediments by USGS, USPHS, and ORNL including radionuclides associated with organic materials.

4. Studies of radioactivity associated with suspended sediments in the river water, including separation of the suspended matter into inorganic and organic fractions.

5. Joint work by the several subcommittees in developing recommendations for river monitoring programs in the future. Public relations as well as legal requirements should be considered.

C. Aquatic Biology. – In presenting the report the chairman, S. I. Auerbach, commented that a major function of this subcommittee was to furnish data for use by the Subcommittee on Safety Evaluation. For completion of Phase I of the Clinch River Study the data recommended were:

1. Results of determinations of radionuclides in fish samples already collected or proposed for completion of the program.

2. Information concerning the fish catch and consumption of fish from the TVA reservoirs, particularly downstream from ORNL.

3. An additional collection of rough fish and recomputation of data from some earlier fish analyses.

4. Further investigation of the effects of dilution factors in the Clinch and Tennessee Rivers, respectively.

It was reported that the subcommittee was continuing its study of the uptake of ^{90}Sr by white crappie through Phase I and overlapping into Phase II of the Clinch River Study. Efforts were being continued to find reliable biological indicators of radioactivity in natural waters.

For Phase II of the study the following objectives and problems were outlined:

1. Evaluation of a biological indicator, e.g., the white crappie.

2. Some study of the effects of harvesting all species of fish, including shad, and processing together for fish meal and cat food (being done at a plant in Clinton, Tennessee).

3. Study of organic detritus, including the concentrations of radionuclides and also use of this material for food by organisms, i.e., entry into the food chain.

4. Algae productivity in the river after the Bull Run Steam Plant is in operation.

D. Safety Evaluation. - The subcommittee reported discussions of the significance of: (1) short-lived radionuclides that decay away in the samples of water or sediment before analysis; and (2) long-lived radioisotopes (for example, tritium) which are not detected by the analytical methods used. It was agreed that the subcommittee should mention these potential contaminants in its report to show that it was aware of the problems and indicate whether there could be important health hazards from these sources.

After discussion it was the consensus of the Steering Committee that the Subcommittee on Safety Evaluation should consider data on radionuclides upstream from White Oak Dam because releases of materials from above the dam might affect the levels of contamination in the rivers downstream.

The subcommittee had considered data on radiation levels in sloughs which had been presented in a progress report. There appeared to be no hazard from this source and no further study of sediments in sloughs was recommended.

Other Discussions and Actions. - The Steering Committee considered two other items of business, namely: future reports on the Clinch River Study, and arrangements for a special meeting with the Harvard Water Resources Group.

After considering various possibilities for future reports, the Steering Committee decided that: (1) the remainder of the study should be covered by one or more status reports, (2) specific investigations conducted during Phase II should be reported in topical reports or included in a status report, and (3) a comprehensive final report and a summary report of a more popular nature should be issued after the conclusion of the entire study.

It was decided that Status Report No. 5 (the present report) should be prepared on the basis of material already submitted, and that further details should be made available in supplements to Status Report No. 5 issued separately. The remainder of Phase I and any important results from Phase II of the study should be covered in

Status Report No. 6, the final status report of the series.

The Steering Committee agreed that the final report should be carefully prepared and attractively published. It was the consensus that this final report should be in two parts: (1) a comprehensive technical report, and (2) a popular-style, widely-distributed summary report. The preparation of these two reports and the extent of distribution were discussed. It was decided that the comprehensive report should be prepared first and that the form and distribution of the summary report, based on the comprehensive report, should be considered later.

The Steering Committee meeting was concluded with the general understanding that at a convenient time a special meeting would be held with Professor H. A. Thomas, Jr. of the Harvard Water Resources Group. The purpose would be discussion of an earlier proposal that the Clinch River Study data be used by the Harvard group for a systems analysis of Clinch River disposal of radioactive wastes.

WATER SAMPLING AND ANALYSIS

Eight months after the beginning of the Clinch River Study in January 1960 the Subcommittee on Water Sampling and Analysis was established (September 22, 1960). A comprehensive plan for collection and analysis of water samples during the study, prepared by this subcommittee and adopted by the Steering Committee, was described in detail in Status Reports No. 1 and No. 2.[1,2] Under this plan water sampling was begun in November 1960 and continued to December 1, 1962.

In brief the program included: (1) operation of a basic network of six (later seven) water sampling stations on the Clinch and Tennessee Rivers; and (2) compositing into weekly (or monthly) samples for analysis portions of daily subsamples of water, with each individual subsample volume proportional to the volume of the daily streamflow passing the particular sampling station.[1,2] From analyses of samples so composited the weekly or monthly mean concentration of each radionuclide or nonradioactive constituent in the water could be determined; and by combining concentrations and streamflows, the total cumulative load of particular constituents passing each station was computed. The program also included special collections and analyses of water samples as needed.

The basic network of stations at which water samples were collected in this study included the following locations:

(1) Clinch River at Oak Ridge Water Plant (CRM 41.5).

(2) White Oak Creek at White Oak Dam (WOCM 0.6).

(3) Clinch River at Gallaher Bridge (CRM 14.4) -- sampling was begun January 8, 1962.

(4) Clinch River above Centers Ferry (CRM 5.5).

(5) Tennessee River at Loudon, Tennessee (TRM 591.4).

(6) Tennessee River at Watts Bar Dam (TRM 529.9).

(7) Tennessee River at Chickamauga Dam (TRM 471.0).

Sampling procedures at each station in the basic sampling network were explained in detail in Status Report No. 2,[2] except for the station at Gallaher Bridge (CRM 14.4). Here sampling equipment was installed in the pump house for the ORGDP water plant, 100 feet downstream from the old Gallaher Bridge on the right bank of Clinch River. Every 4 hours a 1-gallon subsample of water was collected (in a separate container for each subsample) from the raw water discharge line of a specially installed sampling pump and intake pipe. Forty-two subsamples of the 1-gallon samples were composited into a weekly sample with the volume of each of the 42 subsamples proportioned to correspond with the discharge in Clinch River at the Scarboro gaging station (and later at Melton Hill Dam) during the sampling period, with time of wave travel between Scarboro (or Melton Hill Dam) and Gallaher Bridge taken into account.

Radionuclide Analyses

As recounted in previous status reports,[1,2,3] 5-gal water samples from the basic network locations were analyzed for radioactive constituents by the U. S. Public Health Service (USPHS) in Cincinnati, Ohio. These analyses consisted of determinations of ^{137}Cs, ^{60}Co, and ^{106}Ru by computations based on gamma spectrometric data and radiochemical separations for ^{90}Sr. Portions of the results of these determinations have been given from time to time in progress reports by the Subcommittee on Water Sampling and Analysis, and in status reports issued by the Steering Committee.[2,3]

Due primarily to continued improvements in the "electronic computer" analysis of the gamma spectrums of cesium-137, cobalt-60 and ruthenium-106, considerable revision of previously reported concentrations of these radionuclides has been made. The revised data on radionuclide analyses, including strontium-90, have been assembled by the subcommittee in a special report to be issued separately as a supplement to this status report (ORNL-3721, Supplement No. 1).[10] This will include all results on radionuclide determinations from the beginning of the basic-network sampling program in November 1960 through December 1, 1962.

The sampling station at the ORGDP water plant, CRM 14.4, was operated by ORNL from the latter part of 1960 until January 8, 1962. These samples and special samples from other locations were analyzed for radionuclides and stable chemicals by the ORNL Analytical Chemistry Division. Results of most of the analyses made at ORNL were given in Status Report No. 2.[2]

Stable-Chemical Analyses

The detailed data from stable-chemical analyses of samples from the basic network stations and the stable-chemical analyses made at ORNL have been reported in Status Reports No. 2 and No. 3,[2,3] in the Subcommittee's Progress Report No. 3,[8] and in and 1961 reports by the Tennessee Stream Pollution Control Board.[11,12] Altogether, the analyses included in the above reports covered two full years of systematic sampling at the original 6 basic network stations, December 1, 1960 to November 30, 1962 and 11 months at the Gallaher Bridge station (CRM 14.4), January 8 to November 30, 1962. The results showed no significant increase in either the various forms of nitrogen or in phosphates between the upper and lower Clinch River stations, and no extreme variations in river conditions during different periods of time.

Since the detailed analytical data would be very voluminous to publish and difficult to use, the stable-chemical analyses for the period July 1961 through November 1962, given in the subcommittee's Progress Report No. 3,[8] are not included in this status report. Instead, in the following section is shown a statistical analysis of all of the stable chemical data for the several stations, including analyses by the Tennessee Stream Pollution Control Laboratory in Nashville and also stable-chemical analyses made at ORNL. It is believed that the statistical parameters will be more useful than a tabulation of the detailed data in defining conditions in the rivers.

The water samples were analyzed for stable chemical constituents in the laboratory of the Stream Pollution Control Board, Tennessee Department of Public Health (TDPH) in Nashville. The purpose of the program was to determine the effect of the stable chemical composition of the river waters upon the fate of radionuclides released to the rivers. Samples analyzed for stable chemical composition were aliquots of composite samples from which portions were also submitted for radionuclide analysis.

The large number of water samples collected and analyses performed have provided a more or less continuous picture of the stable chemical composition of the two rivers for approximately two years. Several computer programs were prepared and used to speed the processing and analysis of the large volume of data from the sampling program.

Tables 1 and 2 contain statistical summaries of the analyses for the period of record. The means which are listed in the tables have been discharge-weighted in order to make them consistent with the discharge-weighting of individual samples.

Table 1. Summary of Stable Chemical Analyses of Clinch River Water[A]

	Clinch River Mile 41.5, 24 Months[C]				Clinch River Mile 14.4, 58 Weeks[H]					Clinch River Mile 5.5, 104 Weeks[I]				
	Mean	Stand.[D] Dev., Monthly	Min.	Max.	Mean	Stand.[E] Dev., Weekly	Stand.[D] Dev., Monthly	Min.	Max.	Mean	Stand.[E] Dev., Weekly	Stand.[D] Dev., Monthly	Min.	Max.
Turbidity, ppm	28	19	6	68						17	16	10	1	86
Apparent color, ppm	197	124	42	542						114	97	50	3	480
Centrifuged color, ppm	20	15	0	62						20	20	14	0	102
pH			7.1	8.2			0.14	7.0	8.1			0.17	7.2	8.5
HCO_3, ppm	117	7.41	105	130	119	11.3	6.08	87	141	112	13.4	7.86	57	135
Acidity, as $CaCO_3$, ppm	3	3	0	10						4	3	2	0	14
Hardness, as $CaCO_3$, ppm	107	17.4	78	148						106	23.8	19.7	69	169
Ca, ppm	27	4.3	18	36	21	2.4	1.8	17	27	27	5.2	3.8	17	43
Mg, ppm	9.4	2.8	5.1	18.0	7.7	1.0	0.71	4.5	10.0	9.4	3.7	2.8	4.1	22.6
Cl, ppm	5	2	1	11	1.6	0.53	0.30	1.0	4.5	3	1	0.9	1	8
SO_4, ppm	12	5	2	23	10	4	1.5	1.0	27	12	5	3	0	29
NO_3, ppm	1.0	0.59	0.2	2.4	2.7	4.0	3.0	0.3	19	1.5	1.7	2.2	0.0	12.7
Fe, ppm	3.4	2.3	0.3	9.2	0.06	0.08	0.06	0.01	0.37	1.7	1.3	0.80	0.1	7.1
PO_4, ppm	0.2	0.09	0.1	0.4	0.22	0.17	0.10	0.05	0.75	0.1	0.1	0.07	0.0	0.6
K, ppm	1.7	0.49	0.8	3.1	1.3	0.20	0.13	1.1	2.3	1.6	0.55	0.42	0.6	3.5
Na, ppm	2.3	1.1	1.0	5.2	2.4	0.50	0.39	1.8	4.7	2.4	1.3	0.90	0.0	9.5
Si, ppm	2.9	0.69	1.5	4.0	1.5	0.72	0.68	0.1	2.4	2.7	0.63	0.42	0.9	4.4
Specif. cond., μmhos/cm	195	36.5	119	263	216	14.4	9.98	190	282	196	42.8	36.5	105	312
Suspended solids, ppm	185	124	18	557	25.3	20.3	14.5	1.0	104	55	46	32	2	275
Dissolved solids, ppm	125	27	67	201	129	20.7	8.43	90	218	133	64	37	25	549
Total solids, ppm	310	121	140	677	154	23.6	14.9	127	231	188	77	49	112	601
Kjeldahl N, as N, ppm	0.7	0.2	0.3	1.3						0.5	0.3	0.2	0.2	1.7
Mn, ppm	0.4	0.3	0.0	1.1						0.1	0.1	0.08	0.0	0.6
Cr, ppm	0.02[F]	0.05[F]	0.00[F]	0.19[F]						0.01[J]	0.03[J]	0.03[K]	0.00[J]	0.18[J]
Sr, ppm[A]	0.073[G]	0.0068[G]	0.058[G]	0.086[G]	0.070	0.0090	0.0067	0.040	0.080	0.069	0.0076	0.0055	0.043	0.080
Discharge, cfs[B]	5,090	2,780	2,610	14,280	4,620	2,525	1,680	545	15,990	5,580	3,853	3,060	359	19,000

[A]Chemical analyses of samples from CRM 14.4, and of samples from all stations for Sr, by ORNL on *filtered* samples. Chemical analysis of all other samples by Tennessee Stream Pollution Control Board on *raw* samples.

[B]Not discharge-weighted. Discharge data is time-weighted.

[C]Sampling period, December 1960 through November 1962.

[D]Standard Deviation of discharge-weighted monthly samples from discharge-weighted mean for sampling period.

[E]Standard deviation of discharge-weighted weekly samples from discharge-weighted mean for sampling period.

[F]Sampled for 19-month period only, May 1961 through November 1962.

[G]Sampled for 42-week period, March 19–25, 1961 through December 31–January 6, 1962. Maximum, Minimum, Mean, and Standard Deviation are for *weekly* samples.

[H]Sampling period, November 28–December 4, 1960 through January 3–8, 1962. Maximum, Minimum, and Mean are for *weekly* samples.

[I]Sampling period, November 27–December 3, 1960 through November 25–December 1, 1962. Maximum, Minimum, and Mean are for *weekly* samples.

[J]Sampled for 85-week period only, April 16–22, 1961 through November 25–December 1, 1962.

[K]For 19-month period only, May 1961 through November 1962.

[L]Sampling period, August 1960 through November 1962.

Table 2. Summary of Stable Chemical Analyses of Tennessee River Water[A]

	TRM 591.4				TRM 529.9				TRM 471.0			
	28 Months[L]				24 Months[C]				24 Months[C]			
	Mean[B]	Stand.[B] Dev., Monthly	Min.[B]	Max.[B]	Mean	Stand.[D] Dev., Monthly	Min.	Max.	Mean	Stand.[D] Dev., Monthly	Min.	Max.
Turbidity, ppm	14	11	3	52	6	7	1	29	7	9	1	35
Apparent color, ppm	88	58	34	272	53	49	5	226	59	52	8	223
Centrifuged color, ppm	23	21	5	95	24	23	0	108	31	27	5	118
pH			7.1	8.2			7.2	7.9			6.6	8.2
HCO_3, ppm	66	10	50	95	70	7.6	52	82	63	8.8	44	87
Acidity, as $CaCO_3$, ppm	3	2	0	8	3	2	1	10	3	2	0	5
Hardness, as $CaCO_3$, ppm	75	18	49	138	75	18	47	127	70	17	44	115
Ca, ppm	21	4.7	13	34	20	4.5	14	30	19	4.3	12	29
Mg, ppm	5.5	2.8	2.7	15.8	5.8	2.8	1.7	13.1	5.5	2.1	2.4	10.2
Cl, ppm	20	9	5	39	15	7	5	32	13	5	6	24
SO_4, ppm	11	4	1	19	12	3	8	21	12	5	8	32
NO_3, ppm	1.8	1.9	0.4	10.4	1.6	2.7	0.4	14.4	1.5	3.1	0.0	16.3
Fe, ppm	1.0	0.75	0.3	3.4	0.5	0.5	0.0	1.9	0.6	0.6	0.1	2.1
PO_4, ppm	0.2	0.2	0.0	1.1	0.2	0.1	0.0	0.4	0.1	0.08	0.0	0.4
K, ppm	1.3	0.41	0.5	2.5	1.8	0.84	0.8	4.8	1.3	0.33	0.7	2.0
Na, ppm	9.5	3.7	3.9	16.0	6.8	2.7	2.9	12.0	5.8	2.4	1.7	11.3
Si, ppm	3.5	0.54	2.4	4.7	3.1	0.48	1.8	3.8	3.4	0.38	2.6	4.1
Specif. cond., μmhos/cm	170	31.9	122	247	177	33.6	115	247	162	26.9	128	221
Suspended solids, ppm	22	13	0	65	15	13	1	43	9	12	1	44
Dissolved solids, ppm	121	27	75	177	112	19	79	149	101	19	71	131
Total solids, ppm	142	25	105	189	126	18	83	156	111	17	75	138
Kjeldahl N, as N, ppm					0.5	0.2	0.3	1.1	0.4	0.2	0.2	1.1
Mn, ppm	0.1	0.1	0.0	0.2	0.0	0.02	0.0	0.1	0.0	0.05	0.0	0.2
Cr, ppm												
Sr, ppm[A]	0.063	0.014	0.039	0.088								
Discharge, cfs[B]	21,420	8,188	13,490	40,790	31,340	14,290	18,870	66,450	38,880	17,400	22,600	76,820

[A]Chemical analyses of samples from CRM 14.4, and of samples from all stations for Sr, by ORNL on *filtered* samples. Chemical analysis of all other samples by Tennessee Stream Pollution Control Board on *raw* samples.

[B]Not discharge-weighted. Discharge data is time-weighted.

[C]Sampling period, December 1960 through November 1962.

[D]Standard deviation of discharge-weighted monthly samples from discharge-weighted mean for sampling period.

[E]Standard deviation of discharge-weighted weekly samples from discharge-weighted mean for sampling period.

[F]Sampled for 19-month period only, May 1961 through November 1962.

[G]Sampled for 42-week period, March 19–25, 1961 through December 31–January 6, 1962. Maximum, Minimum, Mean, and Standard Deviation are for *weekly* samples.

[H]Sampling period, November 28–December 4, 1960 through January 3–8, 1962. Maximum, Minimum, and Mean are for *weekly* samples.

[I]Sampling period, November 27–December 3, 1960 through November 25–December 1, 1962. Maximum, Minimum, and Mean are for *weekly* samples.

[J]Sampled for 85-week period only, April 16–22, 1961 through November 25–December 1, 1962.

[K]For 19-month period only, May 1961 through November 1962.

[L]Sampling period, August 1960 through November 1962.

It will be noted from the tables that the stable inorganic chemical composition of each of the two rivers has been fairly constant, except for rather wide variations in the suspended sediments and a few other related constituents. Tennessee River water contains somewhat less calcium, magnesium, bicarbonate, and suspended and dissolved solids, and somewhat more sodium and chloride, than dose the Clinch River water. In the water of both rivers, however, the predominant constituent is the anion bicarbonate, and the predominant cation is calcium. At downstream stations the concentration of suspended materials is generally less than at upstream stations.

Statistical comparisons of variations in concentration between constituent pairs were produced by one of the computer programs used in the analysis of the stable chemical data. Correlation and regression coefficients, and squares of the standard errors of estimate, were calculated for chemical constituent pairs for each of the sampling stations. Correlations were considered to be significant if the correlation coefficients were significantly different from zero at the 99.7% confidence level and had relatively small standard errors of estimate.

Certain gross interpretations can be suggested on the basis of the calculated correlation coefficients.[13] Turbidity, apparent color, and centrifuged color have a positive correlation with suspended solids, as expected. For example, the correlation coefficient obtained from a comparison of variations in the turbidity with variations in the suspended solids content of 24 weekly water samples from CRM 41.5 is 0.76. This is well above the value 0.47 which would indicate a probability of 99.7% that the correlation is significant. Iron and manganese, and at some stations silica and potassium, appear also to vary in the same manner as the suspended material in the water, suggesting that those constituents are present in the water primarily as solid particles.

In the Tennessee River, the specific conductance of the water appears to vary directly with the content of the major ionic constituents in the water, with notable exceptions of calcium and bicarbonate. These two constituents do not appear to be related to specific conductance in the Clinch River either. This might be taken to indicate that appreciable amounts of these two ions are in the water in the form of suspended particulate matter.

The suspended sediment content, turbidity, and color of Tennessee River water vary directly with discharge, but this pattern is not apparent in Clinch River water. At two stations, stable strontium shows a positive correlation with calcium, as one might expect if the two chemically similar elements were both derived from natural weathering processes. In the lower Clinch River, however, stable strontium varied inversely with suspended solids, suggesting that the strontium may have been associated with solids which were removed from the water during sedimentation, or was diluted by runoff with low strontium content.

Extraneous Sources of Release of Radionuclides to Clinch River

In portions of the Clinch River Study that involve radionuclide loads in the river, calculations of the loads are based on the assumption that the only significant releases of radioactive liquids to the Clinch River are the monitored discharges through White Oak Dam. It is desirable to occasionally provide assurance that no other release points exist. A four-phase investigation for this purpose was conducted during 1963.[14]

The gates in White Oak Dam were closed on April 8, 1963, and fluorescent dye was mixed with White Oak Lake water along the upstream face of the dam. After 24 hours, water samples for fluorescent analysis were collected from very small seeps and small puddles on the creek overbank immediately downstream from the dam. No fluorescent dye was detected in the samples. However, flow in the small seeps appeared to have increased slightly and the creek overbank had become more wetted as a result of the higher lake level, which suggested a minor amount of seepage through the earth-fill dam.

On March 1, 1963, a visit was made to the Tower Shielding Facility, which is the only ORNL facility outside the White Oak Creek drainage basin from which radionuclides might enter the Clinch River. It was learned that the reactor cooling water at this facility normally circulated in a closed system. When radioactive liquids were to be released from the reactor they were piped into a large closed tank, and later transferred to a tank truck and transported to the ORNL area within the White Oak Creek basin for disposal. This arrangement appeared to preclude unmonitored contamination of the river from the Tower Shielding Facility.

Several water samples were collected on February 21, 1963 from small streams draining areas adjacent to the White Oak Creek drainage basin and from the two streams that drain the Y-12 area. Sample locations are shown in Fig. 1. Gross-gamma counting of the samples showed no radioactivity above background.

The data collected in the surveys described above indicate little possibility that significant amounts of radioactive contamination is introduced into the Clinch River through surface drainage at any place in the Oak Ridge reservation except White Oak Dam and

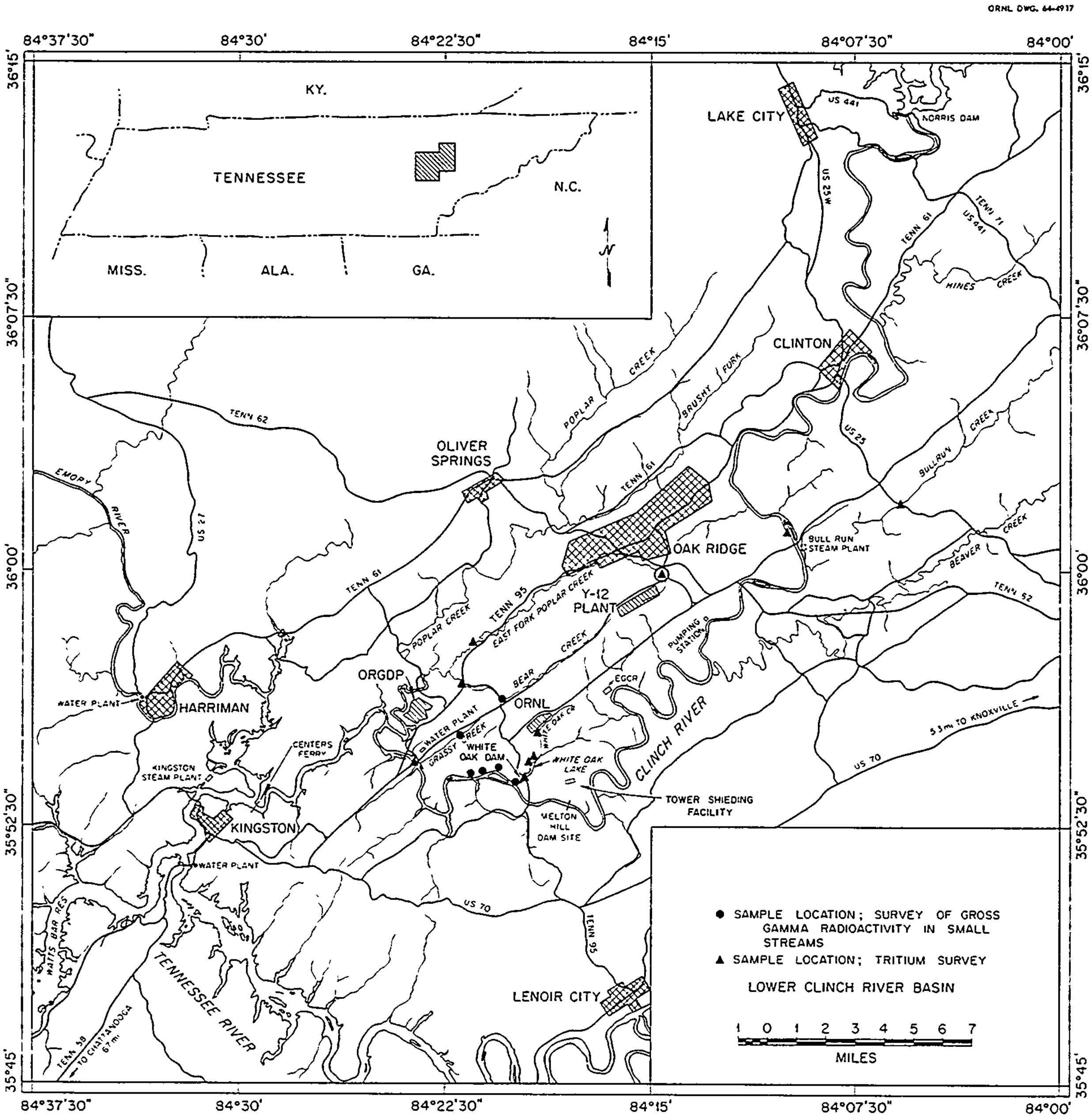


Fig. 1. Sampling Locations for Surveys of Gross Gamma Radioactivity and Tritium in Streams, Oak Ridge Area

the ORGDP area. Drainage from ORGDP is either inside the security fence or is affected by Clinch River waters, and thus could not be readily checked. However, it is known from the operations at ORGDP that gross quantities of fission products are not handled at that plant.

On February 15, 1963, ten water samples were collected from the area in and around the Oak Ridge reservation and submitted to the U. S. Geological Survey Tritium Laboratory in Washington, D. C. for determination of their tritium contents. Four of the samples were collected from sources in the White Oak Creek basin known to contain radioactive wastes. These sources were: the effluent from the process waste water treatment plant at ORNL; Well No. 57 in the waste disposal pit area; water from the weir west of the waste pit area which included surface seepage west of the area; and water from White Oak Lake at White Oak Dam. Analyses of these samples showed the tritium contents given in Table 3. The results show that tritium was present in the wastes sampled and in White Oak Lake water.

Six water samples were collected from four streams in the Oak Ridge area outside the White Oak Creek basin in order to obtain a background value for the tritium content of streams in the area. Samples were collected from the Clinch River at CRM 48.2 and 14.4, Bull Run Creek at Highway 25W, East Fork of Poplar Creek at Pine Ridge and at the gaging station near its mouth, and Bear Creek at the gaging station near its mouth. The results of the analyses of these samples are shown in Table 4. The reason for the rather high "background" concentration of tritium in the two samples from the Clinch River is not known; and the source of the even higher tritium content of Bear Creek downstream

Table 3. Tritium Analyses of Water Samples Contaminated with Radioactive Wastes

Source	Concentration: Tritium Units*	Concentration: pc/l
Process Waste Water Treatment Effluent	105,000	336,000
Well No. 57 in Waste Pit Area	675,000	2,160,000
Weir West of Waste Pit Area	833,000	2,666,000
White Oak Lake at White Oak Dam	43,000	138,000
Maximum Permissible Concentration (water):		
MPC_w (continuous occupational exposure)	0.93×10^7	3×10^7

*Definition of Tritium Unit: A tritium unit (1TU) corresponds to a concentration ratio of 1 atom of tritium (^3H) to 10^{18} atoms of hydrogen ($^1H + {}^2H + {}^3H$). There is a tendency now to call this value the "Tritium Ratio" rather than "Tritium Unit".

The concentration of tritium in water is easily converted from tritium units to curies per volume; i.e., multiply the concentration in TU's by 3.2 to convert to picocuries ($\mu\mu c$) per liter (see last column in table above, and Table 4 below.

Table 4. Tritium Analyses of Water Samples from Streams in Oak Ridge Area

Source	Concentration: Tritium Units	Concentration: pc/l [a]
Bear Creek at gage	1110	3550
Clinch River at CRM 48.2	430[b]	1375
Clinch River at CRM 14.4	530[b]	1695
East Fork of Poplar Creek at Pine Ridge	287	920
East Fork of Poplar Creek at gage	244	780
Bull Run Creek at Highway 25W	230	735

[a] See footnote under Table 3.

[b] Average of duplicate analyses; difference is approximate contribution from White Oak Lake.

from the Y-12 plant has not been determined. The higher concentration in Clinch River at CRM 14.4 than at CRM 48.2 may be due to tritium discharged to the river through White Oak Creek.

RADIOACTIVITY IN BOTTOM SEDIMENTS

The various studies of river sediments have taken an unusually large proportion of time and effort in the Clinch River Study during the period covered by this Status Report (April 1962 to December 1963). Sediment investigations, which are of basic importance in the river study, have included review of much existing data from TVA surveys of sediment deposition, results from extensive core-sampling of river sediments, data from laboratory analyses of the cores, experimental studies of the behavior of radionuclides in sediments, and special investigations to define relevant factors in the incorporation and retention of radionuclides in sediments.

Most of the sediment studies mentioned above are reported in the several subsections below. Only a summary is given of a voluminous report on data evaluation which was prepared by the Subcommittee on Bottom Sediment Sampling and Anslysis. Also the data from specific laboratory analyses of cores of Clinch River sediments collected during the summer of 1962 are not included. It is planned that the report on data evaluation, mentioned above, and the analytical data from the core analyses will be published separately in a two-part supplement to this status report (ORNL-3721, Supplement No. 2, Parts A and B, in preparation).[15, 16]

Summary of Report on Data Evaluation

A report evaluating the data available in January 1963 pertaining to radioactivity in sediments of the Clinch and Tennessee Rivers was prepared by the

Subcommittee on Bottom Sediment Sampling and Analysis. The subjects considered in this report included: sediment deposition, distribution of radioactivity in time and space, downstream limits of detection, physicochemical characteristics, sampling tools, retention of radioactivity in bottom sediments, radiation dosage, and recommendations. Credit for the data and reports used in this evaluation was accorded to personnel of the ORNL Applied Health Physics Section, ORNL Waste Disposal Research Section, U. S. Public Health Service, U. S. Geological Survey, and the Tennessee Valley Authority. A summary of the subcommittee's report, including the recommendations, which was presented at the meeting of the Steering Committee in Cincinnati, Ohio on February 6, 1963, is given below, (pages 29 to 41).

Factors that Influence Radionuclide Concentration

Of the many physicochemical factors that may influence the deposition and retention of radioactive materials in bottom sediments of the Tennessee River system, a few have been observed specifically though not fully defined. Two important factors that have been identified are: (1) annual variations in the radionuclide loads released through White Oak Dam, and (2) channel geometry. Other significant factors which have been investigated are mineralogy, particle size, water depth, and exchangeable calcium in the sediment.

Radionuclide Loads Released.-- Data on the total quantities of radionuclides released through White Oak Dam have been compared with data on concentrations of radionuclides at the surface of bottom sediments downstream during concurrent periods. These data indicate that concentrations in the sediments vary directly with changes in the radionuclide loads released. This is illustrated in Fig. 2 (page 31) by comparing the concentration of ^{137}Cs in sediments at CRM 1.1 to the annual releases of this radionuclide through White Oak Dam. From records furnished by the ORNL Applied Health Physics Section, similar relationships have been found for ^{90}Sr, ^{106}Ru, and ^{144}Ce at CRM 1.1.

Plots of data for other observation sections in the Clinch and Tennessee Rivers have shown relations similar to those for CRM 1.1 illustrated in Fig. 2. These relations could be explained by either of two hypotheses regarding the incorporation of radionuclides in the bottom sediments. These hypotheses are: (1) that radionuclides are sorbed on suspended sediments prior to their release through White Oak Dam and the annual samples of bottom sediments are representative of sediment deposited during the entire preceding 12 months; and (2) that the radionuclides are released through White Oak Dam as ions in solution and that variations in the amount of a particular radionuclide released cause similar variations in its concentration in the river water and in the amount sorbed *in situ* by the bottom sediments.

The first hypothesis, "(1)" above, presupposes regular, continuous deposition of sediment during the 12 months preceding sampling. The second hypothesis, "(2)" above, presupposes rapid establishment of chemical equilibrium between the water and sediment. It, too, presupposes regular, continuous deposition of sediment over the sampling section unless there are fairly constant releases of radionuclides during the year. Without this constancy in releases, each incremental layer of sediment must be covered by other

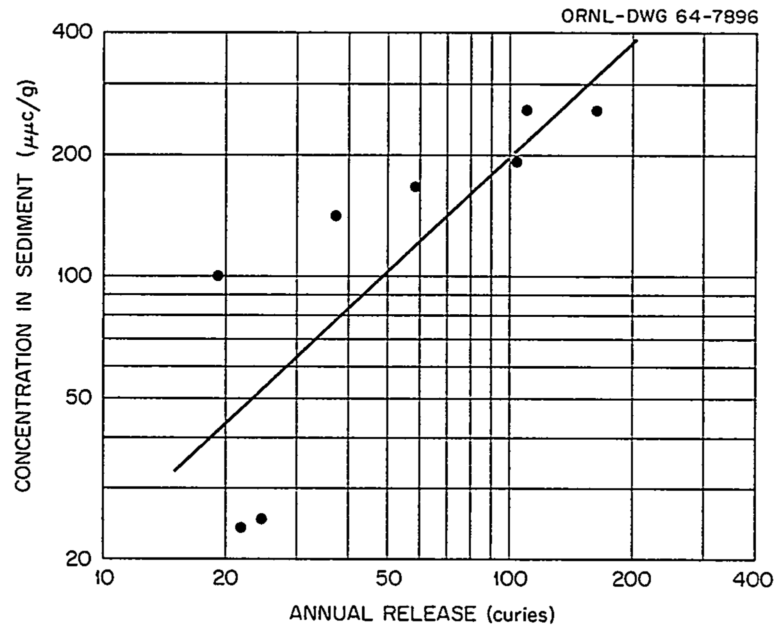


Fig. 2. Comparison of Concentrations of Cesium-137 in Bottom Sediments at CRM 1.1 Determined Annually to Annual Releases of this Radionuclide from White Oak Lake

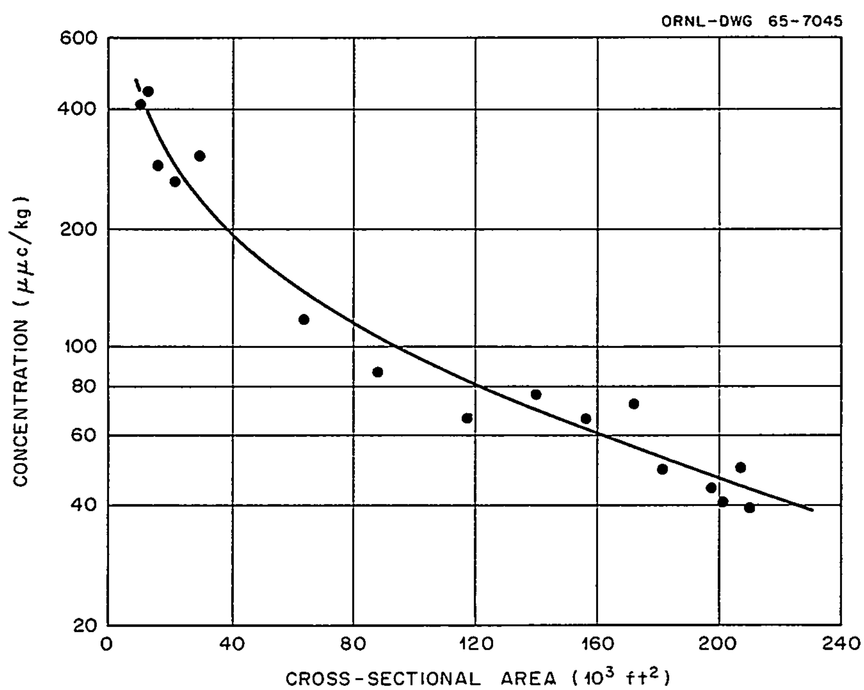


Fig. 3. Variation of Total Identified Radionuclide Concentration in Sediments with Cross-Sectional Area of Flowing Water

sediment and isolated from contact with river water; otherwise the sediment sample taken annually would represent equilibrium only with the radionuclide concentration in the river water just before sampling. In the aggregate of sediment deposition and scour over a period of time, a net increase or net loss in the sediment accumulation at a particular sampling section is not a prerequisite for either hypothesis.

It is known from monthly monitoring data that the amounts of radionuclides released through White Oak Dam have not been constant during the 12-month observation periods. Also, data from TVA sediment surveys indicate that deposition of sediment at the sampling sections is not continuous. The only part of the study reach in which net deposition has occurred in every 5-year sediment-survey period is downstream from the mouth of the Emory River. It must be assumed, therefore, that the sediment removed during periods of scour does not cause net sediment loss over a major width of the section. Pertinent TVA sediment range sections show wide zones of continuous, net accumulation of sediment on the sides of the stream channel, and only limited zones in the deeper parts of the channel which are subject to scouring. Such scouring is probably the result of a few periods of high flow in winter.

*(In the case of ^{137}Cs, it is known that more than 80 percent of the radionuclide in the water passing through White Oak Dam is associated with suspended solids.[8] It is also known that the reaction by which cesium is sorbed on the sediment is not readily reversible (see later section on behavior of nuclides, (pages 52 to 62). It may be assumed,therefore, that the incorporation of ^{137}Cs into bottom sediments conforms to hypothesis (1).

The direct relationship of radioactivity in sediments to the load of radionuclides released is useful in establishing the longitudinal limits of detection of fission products.

Surveys of radionuclides in bottom sediments were extended beyond Guntersville Reservoir to the Ohio in 1961. Based on these surveys, contamination of bottom sediments from the mouth of White Oak Creek to the junction of the Tennessee River with the Ohio River near Paducah, Kentucky was limited to ^{106}Ru. Contamination with ^{137}Cs, ^{90}Sr, ^{144}Ce, and ^{60}Co extended into Pickwick Landing Reservoir; and contamination with ^{95}Zr-^{95}Nb and with ^{90}Y and trivalent rare earths extended into Guntersville Reservoir. In earlier years, the annual releases of ^{137}Cs, ^{90}Sr, ^{60}Co, and ^{144}Ce have been several times higher than in 1961. Thus, their contamination of bottom sediments (observed at the surface of the sediments) may have extended downstream to the Ohio River at some previous time.

Contamination of bottom sediments in the Clinch River has not extended upstream from White Oak Creek except in the three-year period of 1957-59. In these years radiation levels above background were detected at least 0.7 mile but less than 3.3 miles upstream from the mouth of White Oak Creek. In the Tennessee River, no radioactive contamination has been detected at an observation section a short distance upstream from the mouth of the Clinch River.

Channel Geometry.-- Two factors which have important influences on the concentration of radioactive materials in bottom sediments are the surface area of the channel bed and the curvature of the channel. These two factors come under the general term "channel geometry".

W. D. Cottrell, in 1959, noted that the concentration of radioactivity is decreased as the radioactivity is dispersed over a larger area.[17] Decrease in concentration with an increase in surface area can be inferred by the relation shown in Fig. 3, page 31, in which the concentration of radioactivity decreases with an increase in cross-sectional area. In the Clinch and Tennessee Rivers the cross-sectional area of the river channel bears a direct relationship to the surface area of the bottom sediments because the wetted perimeter in most sections increases as the cross-sectional area increases.

The influence of velocity, or more fundamentally of turbulence, must be tacitly recognized in considering the effects of cross-sectional area upon radionuclide concentrations in sediments. In this particular river system, increases in cross-sectional or surface areas coincide with decreases in velocity. Decreases in velocity and in turbulence should permit smaller particles to settle to the stream bed. Cottrell[17] noted the tendency for the radioactivity levels in bottom sediments to be low in the first few miles downstream from a dam (higher velocities) and to increase further downstream in the reservoir (lower velocities). The decrease in concentration immediately downstream from a dam was attributed to bottom scour. The tendency for radioactivity to increase in the downstream direction within a reservoir has been observed repeatedly.

Without considering localized influences, such as variations in flow area within reservoirs and in channel curvature, the general trend in radionuclide concentration in the Tennessee River is a decrease in the downstream direction. The downstream decreases in concentration of cesium, ruthenium, cobalt, and strontium observed in bottom sediments at several points in the Tennessee River have been compared with predicted reductions based on (1) dilution by flow, and (2) dilution by uncontaminated sediments. The observed reduction in concentration of each radionuclide, except ^{90}Sr, was greater than the predicted reduction due to dilution by flow but not as great as the reduction predicted from dilution by uncontaminated sediment. The reduction in the concentration of ^{90}Sr was about equal to that predicted by flow dilution.

The downstream variation of radioactivity in the Clinch River is more complex and both increases and decreases in concentration with distance have been observed. The general trend is for the concentration in sediments to increase in the downstream direction but there is a zone of relatively high radioactivity that seems to center between CRM 8 and CRM 12.

Channel curvature is another aspect of channel geometry which has been found to influence radioactivity in the bottom sediments. From the available data it appears that the concentration of radioactivity in the sediments decreases as curvature increases, and that more deposition of sediment with higher concentrations of radioactivity have been observed in the portions of a section that are on the inside of a bend. Some aspects of the influence of bends on sediment deposits were discussed in an earlier status report.[18]

<u>Physicochemical Properties of Sediments.</u> -- Physicochemical properties of bottom sediments in the Clinch River that have been investigated are size distribution, mineralogy, sorption capacity, and exchange capacity. For these investigations analytical tests have been performed only on composites of cores collected in sections spaced 1 to 3 miles apart. Definite conclusions about the influence of physicochemical properties of the sediments must await more intensive sampling programs, but some preliminary judgments can be formed.

Sorption of radionuclides on river sediments can be expected to increase with decreasing particle size because: (1) the relative content of clay minerals, the mineral group with the highest cation sorption capacity, usually increases as the median particle size of the sediment decreases; and (2) surface area (area per unit weight) increases as particle size decreases and the exchange capacity in many minerals varies directly with surface area of the particles. A relationship between particle size and sorbed radioactivity (^{137}Cs) is shown in Fig. 4. The ^{137}Cs concentration has been adjusted for the influence of the "areal dilution" illustrated in Fig. 3 because the correlation coefficient is more significant if the areal dilution is considered.

The reasons for the generally low ^{90}Sr concentrations in the bottom sediments, and for variations in the stable strontium concentrations, have been suggested by the results of exchange capacity tests. The low strontium concentrations appear to be the result of low exchange capacity of the sediments and the high concentrations in Clinch River water of dissolved calcium, which competes with strontium in exchange reactions. Variations in concentration of ^{90}Sr in the bottom sediments appear to be directly related to the "exchangeable" calcium associated with these sediments. The results of sorption tests, exchange capacity tests, analyses of river water, and pH determinations lead to the conclusion that much of the strontium is associated with, and much of the "exchangeable" calcium is derived from, calcium carbonate incorporated in the sediments rather than calcium ions sorbed by the sediments.

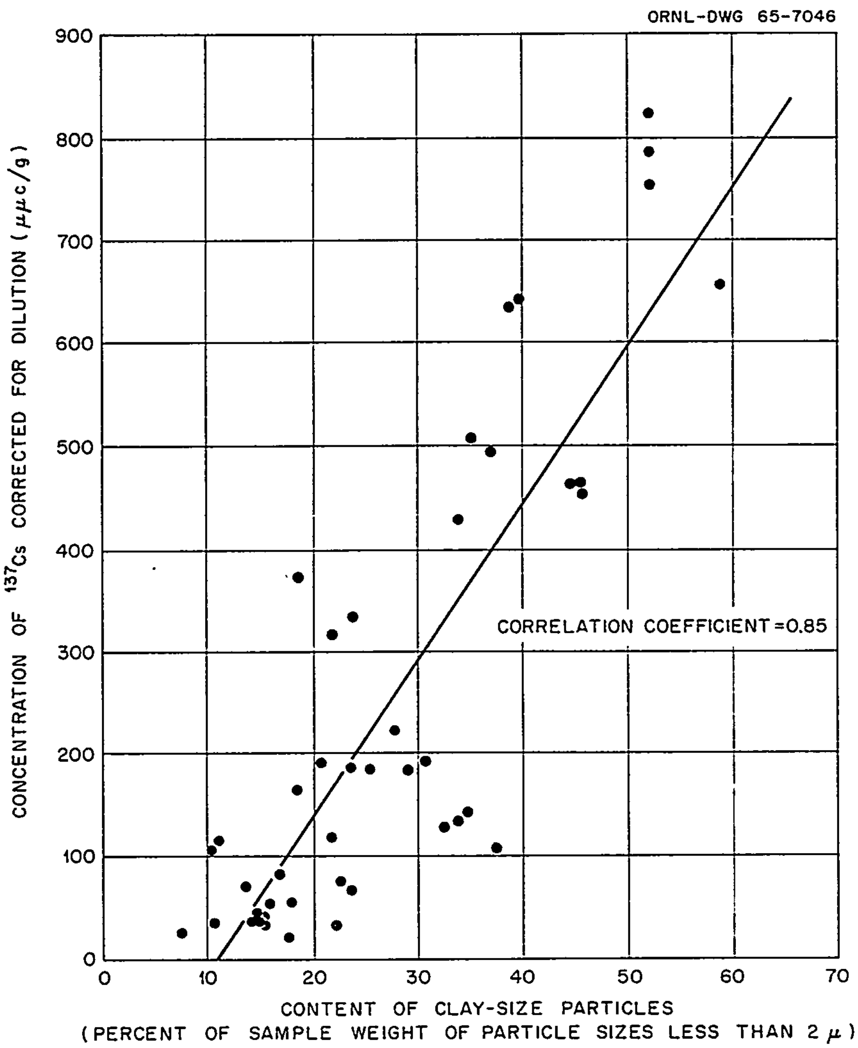


Fig. 4. Relationship between Cesium Concentration and Clay-size Particle Content of Bottom Sediments of Clinch and Tennessee Rivers

Inventory of Radioactivity in Bottom Sediments

Investigations of factors that influence the retention and deposition of radioactivity in bottom sediments have not reached a status that permits an assessment of the total activity held in these sediments. In reaches where information indicates that there has been continuous deposition of sediment, computations to assess this total may be possible, assuming constant rates of deposition and collection of representative dredge samples annually. Such computations would only be feasible in the reach of Watts Bar Reservoir extending from the mouth of the Emory River on the Clinch River to Watts Bar Dam on the Tennessee River. In this reach observations by TVA at the sediment ranges indicate a relatively long-term trend toward continuous accumulation of radioactive sediments. Upstream from this reach, in the Clinch River, alternate periods of gains and losses in sediment deposits have occurred; in the fifteen years that sediment deposition has been measured the gain has been slightly greater than the loss (from records of TVA surveys). Downstream from this reach, in the Tennessee River, the changes in the volume of sediment deposits in Chickamauga Reservoir have been almost negligible, based on review of records furnished by TVA.

In the Clinch River, upstream from the mouth of the Emory River, results of a reconnaissance coring survey did suggest that only a small fraction of the radionuclides released to the river are retained in the channel bed. Based on this survey, made in 1960, there are perhaps 70 curies of fission products in the upper 6-10 inches of sediments between the mouths of White Oak Creek and the Emory River. The coring tool used in this survey did not penetrate the entire thickness of the radioactive sediments; downstream from CRM 14 the thickness of the sediments averaged about 2 1/2-3 feet while upstream the thickness was somewhat less. It appears that with complete penetration the results of such a coring program would indicate a total activity only a few times greater than 70 curies. This total radioactivity in the stream bed is relatively small in comparison with the gross beta activity released and passing down the river, and would indicate a small factor of retention by the sediments. For example, the annual releases to the Clinch River of gross beta activity in the two years 1959 and 1960 were 937 curies and 2190 curies, respectively.

Sediment Sampling Equipment

The coring work in 1960 and other field work provided means for evaluating equipment used in investigations of radioactivity in bottom sediments. For example, the results of the preliminary coring program showed that longitudinal variations in radiation levels were much the same by various methods of determination. However, quantitative estimates of the radionuclide content of the full depth of sediments could not be made because of deficiencies in coring equipment, especially plugging of the sampler and compaction of the core.[19]

Field evaluation of various coring devices showed good penetration and undisturbed samples were obtained if the coring tools were tubes at least 3-1/2 inches in diameter with sharp, smooth cutting edges. Sample retention in these tubes was greatly improved if spring-finger retaining rings were used. A volumetric silt sampler developed by TVA and a Phleger-type coring sampler developed by personnel of ORNL and USGS were found to be useful for full-depth sampling in this evaluation.

For reconnaissance the hand operated dredge (Ekman or Pettersson) was found to be advantageous. However, the usefulness of small dredges in studies of bottom sediments requiring quantitative determinations is limited because the sampling depth for the dredges is difficult to determine, the sample is disturbed, and the small-particle fraction of sample may be lost by washing.

Other equipment considered in the data evaluation report are devices to measure the horizontal distribution of radioactivity in bottom sediments by in-situ measurements of gamma radiation at the surface of the sediments. Most devices used for such measurements perform well in the Clinch River. Further downstream in the Tennessee River the relatively high levels of natural radioactivity and fallout as compared to levels of radioactivity released from ORNL tend to limit the usefulness of these detectors. Garner and Kochtitzky [20] felt that the variations in radiation levels observed downstream from Hales Bar Dam in 1952 were due to the variation in the deposition of naturally occurring radioactive sediments in the river. In 1961, even though fission-product contamination was observed, the variations and magnitude of the radiation levels downstream from Hales Bar Dam were about the same as in 1952.

Efforts to estimate gamma dose rates at the surface of bottom sediments from "Flounder" count rates are discussed in the report. The Flounder instrument is calibrated with a radium source. Definition of the dose rate from the Flounder count rate has proved to be very difficult because of differences in the spectrum of radiation from the radium source and from the river sediments, and also the different and indefinite shapes of the radiation sources in sediment deposits as compared to the calibration source.

Recommendations

The summary of the data evaluation report included definite recommendations for a comprehensive program of investigation of radioactivity in bottom sediments. However, since that time, the program plans have been modified to meet project time schedules and the recommendations originally submitted will not be repeated here in detail.

In brief, the recommendations urged efforts to increase information about the absolute retention of each radionuclide in bottom sediments in the Clinch River. This determination would provide a better understanding of the influence of bottom sediments on the transport of radionuclides through the river, assist in evaluation of the usefulness of streams for the disposal of radioactive materials, and provide a better basis for safety evaluations.

The importance of geochemical studies to delineate the physicochemical processes which lead to the incorporation of radionuclides in bottom sediments was emphasized. It was suggested that the geochemical studies should include investigations of the physicochemical state of radionuclides in bottom sediments and in the river water, of sorption by lithologic biotic sediment, of desorption processes, and of the depth to which chemical reactions occur in the deposited material.

Core Sampling of Bottom Sediments in the Clinch River

Introduction

Numerous undisturbed core samples of bottom sediments in the Clinch River were obtained during the summer of 1962 in order to define the horizontal and vertical distribution of radioactivity in the bed of the river. The Swedish Foil Sampler was used in the 1962 coring program.[21] This sampler, which takes cores slightly less than 3 inches in diameter, was selected as the best tool for obtaining "undisturbed" sediment samples.

It was expected that after the vertical and horizontal distribution of activity had been determined the core samples would provide information on: (1) the concentration of each of the major radionuclides in the bottom sediments at the 14 cross sections sampled and the total amount of each radionuclide present in the study reach; (2) the distribution of major radionuclides and of physicochemical components in the sediments; (3) the chemical form of radionuclides and thereby the mechanisms by which they were incorporated in the bottom sediments; and (4) characteristics of sedimentation in the Clinch River.

The primary purpose of the investigation was to obtain quantitative data concerning the fate of radioactive materials currently being discharged to the river -- the first general objective of the Clinch River Study. Also, the investigation was designed to contribute to the second general objective of the Clinch River Study, namely, to determine and understand the mechanisms of dispersion of radionuclides released to the river.

A brief summary of the coring study and an indication of the results are given below. A more detailed account is being made available in Part B of the supplement to this status report, prepared for separate distribution.[16]

Coring Operations

More than 130 cores were collected from the portion of the Clinch River downstream from CRM 23 during June, July, and August 1962. Results of the coring work were reported at a meeting of the Steering Committee on February 6, 1963.[5]

The Swedish Foil Sampler is a piston-type sampler in which thin axial strips (foils) are used to decrease friction between the sediment core and the sample tube while coring is proceeding. Cores as much as 14 ft in length were obtained in water as much as 45 ft in depth using a drilling tower mounted on a barrel float. The sampling operation was carried out by a contractor (Sprague and Henwood, Inc.) whose personnel were specialists in sampling with this device.

Cores were obtained at 4 to 12 verticals across each of 14 cross sections in the Clinch River, some of which were located at TVA sediment ranges. In addition to the cross sections sampled in the Clinch River, 2 sections in the Emory River and 2 sections in Poplar Creek were sampled. The locations of the sections where coring was done are shown in Fig. 5. Sections were selected for sampling on the basis of the longitudinal variation in radiation levels determined in 1960 and 1961 by personnel of ORNL and USGS. The location of the sampling verticals in each section was based on radiation levels measured _in situ_, and penetrometer probing of sediment depths. These two types of measurements were made immediately before coring.

From 218 coring attempts in the Clinch River, which included a number of "second tries", 136 satisfactory cores were obtained. A total of 27 cores were obtained from Poplar Creek and the Emory River. The location by river mile of each of the sections where coring was done and the number of cores obtained at each station are listed in Table 5.

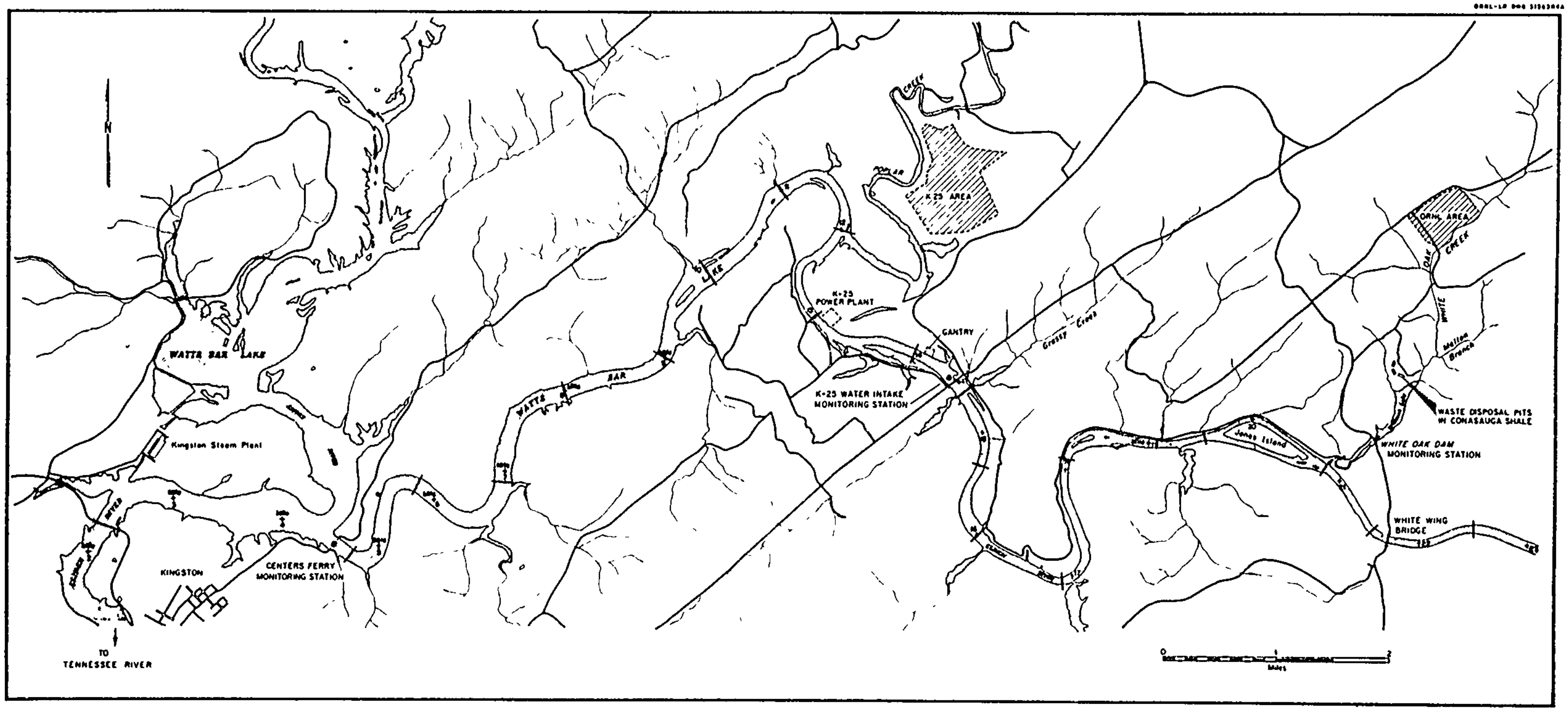


Fig. 5 Map Showing Locations of Sediment Core Sampling Stations

Table 5. Summary of 1962 Sediment Coring Program

Location Mile*		Number of Coring Sites	Number of Cores Taken**	Number of Cores Gross Gamma Scanned	Feet of Core Gross Gamma Scanned
			CLINCH RIVER		
CRM	1.3	10	13	11	37.9
	4.3	12	14	11	33.5
	7.5	9	10	10	50.1
	10.0	10	14	14	52.8
	11.9	10	14	14	58.4
	12.1	11	15	12	53.5
	14.0	5	6	6	38.0
	16.0	7	7	7	20.6
	17.5	6	8	8	40.7
	19.2	5	6	6	20.0
	20.5	8	12	9	17.4
	20.8	5	6	5	13.6
	21.0	4	6	6	22.7
	22.8	5	5	4	23.1
	Subtotal	107	136	123	482.3
			POPLAR CREEK		
PCM	3.1	5	5	5	26.9
	4.5	6	7	7	19.6
	Subtotal	11	12	12	46.5
			EMORY RIVER		
ERM	1.9	7	8	6	11.6
	5.1	7	7	5	13.8
	Subtotal	14	15	11	25.4
	TOTAL	132	163	146	554.2

*Upstream from mouth of stream.
**Includes "second tries".

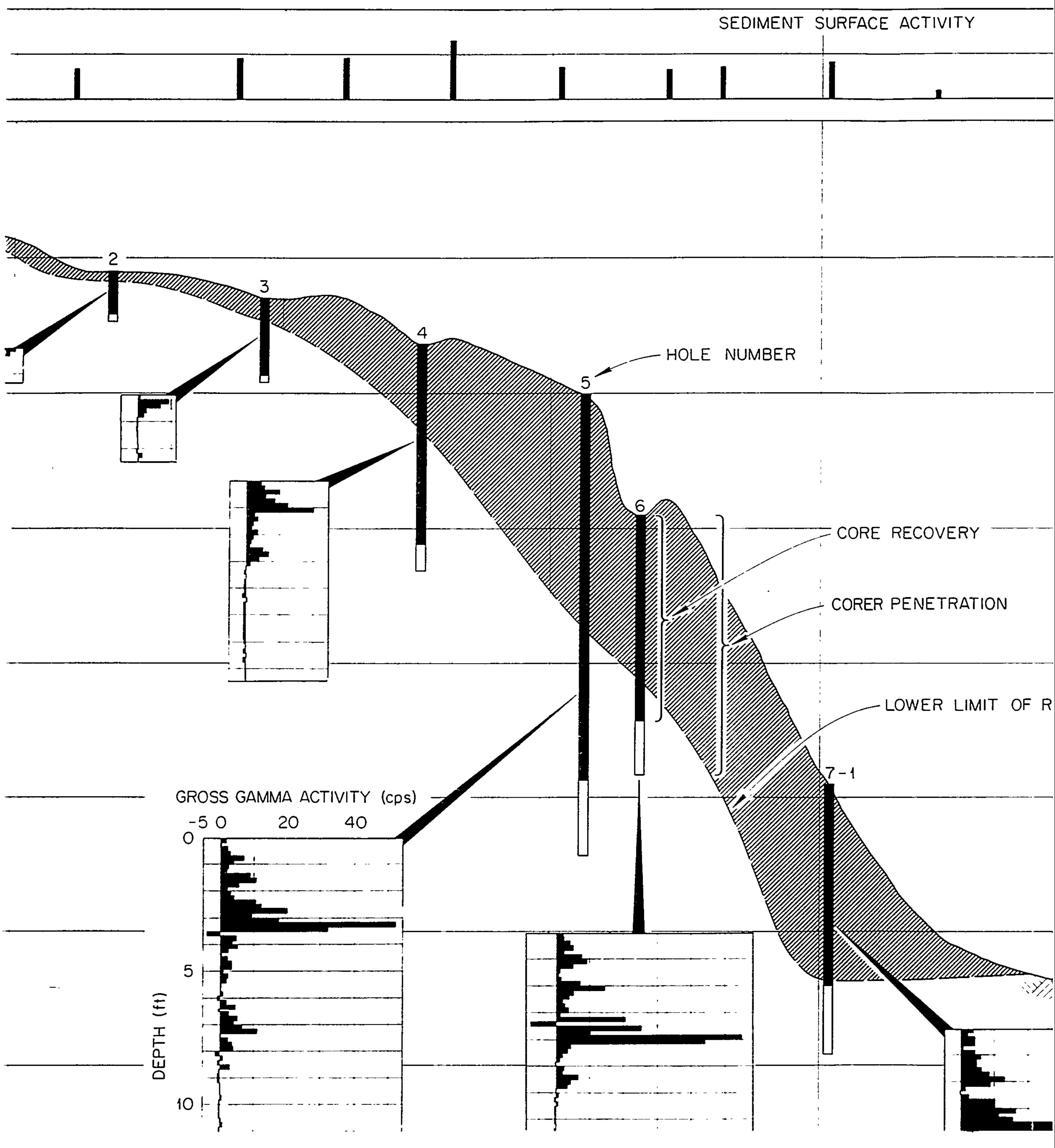
A graphical summary of experience in the coring program at CRM 7.5 is shown in Fig. 6. The cross-sectional shape, lateral distribution of surface radioactivity, depth of radioactive zone, location of core verticals, depth of penetration, and depth of core recovery are included in this figure.

Gross Gamma Scanning of Cores

The first step in the processing of the cores was measurement of incremental variations in gross gamma radioactivity with depth and analysis of the gamma spectra of individual cores for radionuclide concentrations. These measurements were made with the "core scanner" of which a schematic diagram is shown in Fig. 7.

The cores were scanned in 2-inch increments. It should be noted that the effective slit width of the 2-inch-high collimator was somewhat greater than the actual 2-inch dimension of the slit. The effective slit width was determined experimentally with a 3-inch-diameter plane source of radioactivity; and a computer program was prepared by R. P. Leinius of the ORNL Mathematics Division to compensate for the effect of the greater effective slit width in the incremental determinations of radioactivity in the 2-inch segments of core.

An example of the results of the gross gamma scans for cores obtained at CRM 7.5 is included in Fig. 6. The plots of vertical distribution of radioactivity in Fig. 6 show that there are two segments for which the gross gamma count is a negative value. Study of these plots and those for other cross-sections indicates that the negative values occur only if there is a great difference in the uncorrected gross gamma counts between adjacent segments. The failure of the computer program to compensate perfectly for these large-magnitude changes in a few cases is due to the characteristics of the computer program used and not to the physical character of the cores.



SEDIMENT SURFACE ACTIVITY
HOLE NUMBER
2
3
4
5
6
7-1
CORE RECOVERY
CORER PENETRATION
LOWER LIMIT OF R
GROSS GAMMA ACTIVITY (cps)
-5 0
20
40
0
5
10
DEPTH (ft)

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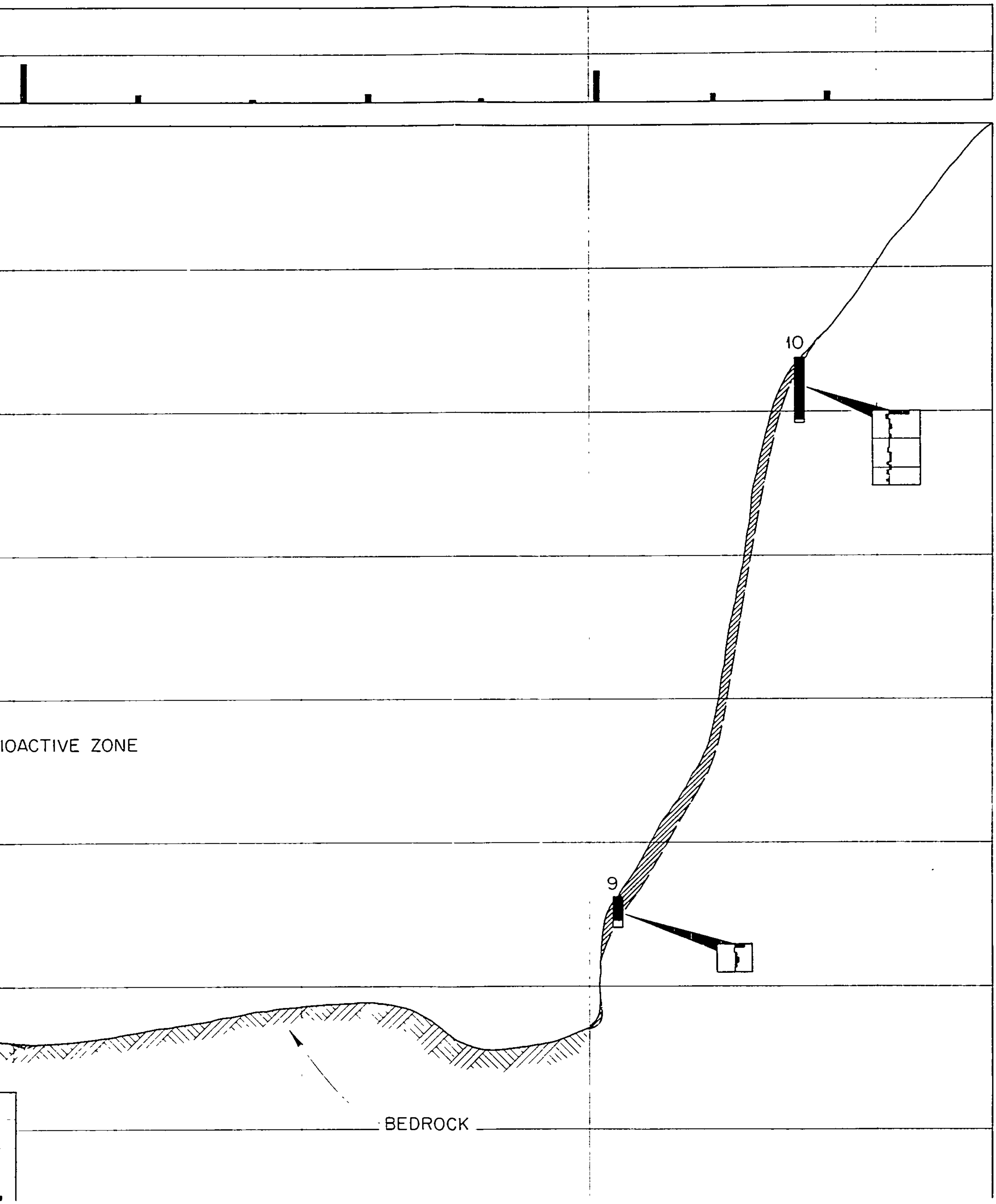

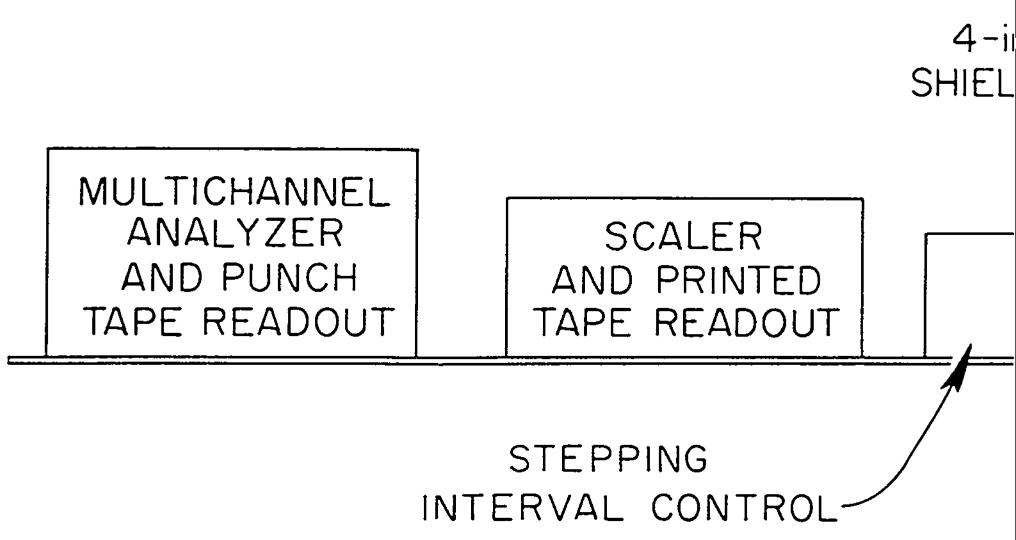
4-i
SHIEL
MULTICHANNEL ANALYZER AND PUNCH TAPE READOUT
SCALER AND PRINTED TAPE READOUT
STEPPING INTERVAL CONTROL

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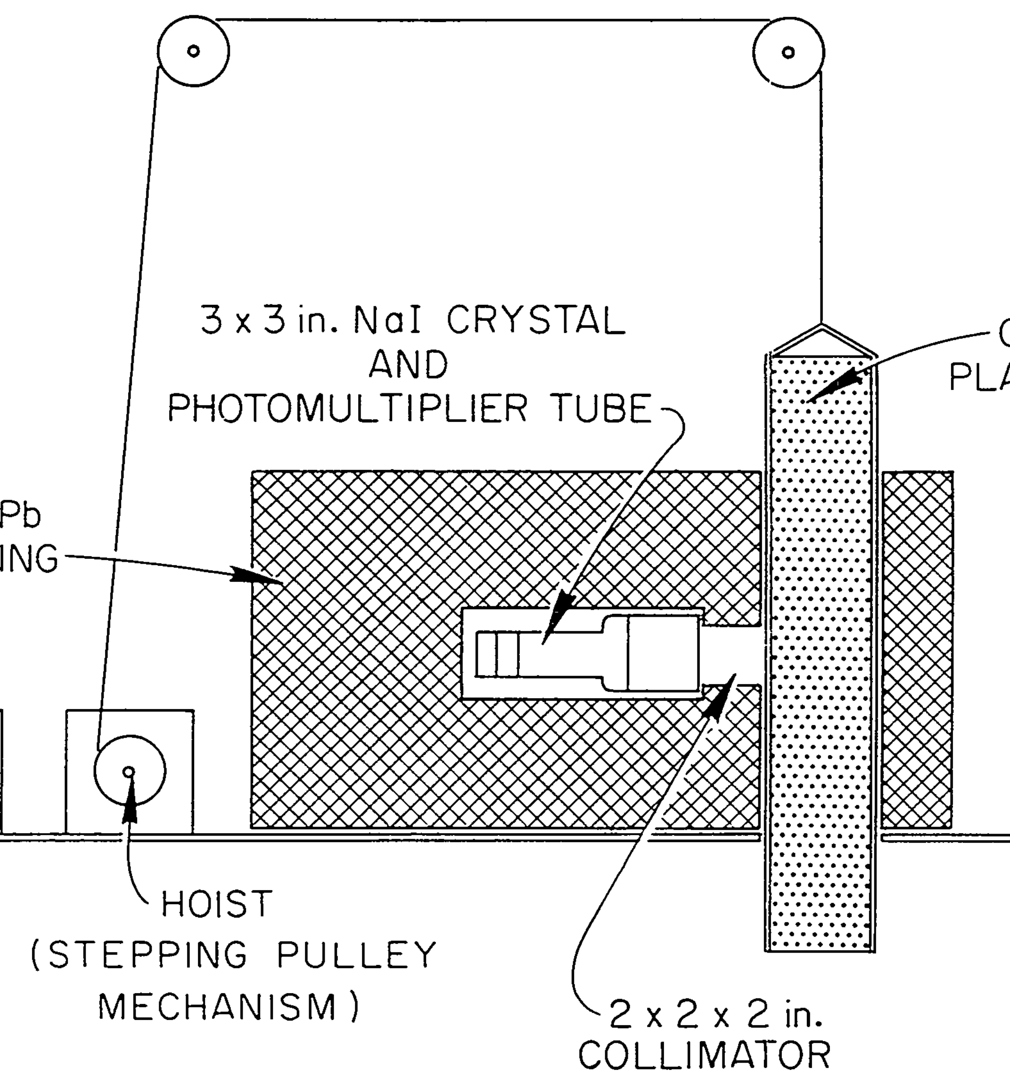

Diagrams similar to Fig. 6 have been prepared for all sections at which core samples were collected during the summer of 1962. Additional diagrams and more detailed discussion of the vertical distribution of radioactivity found in the cores and the results of other analyses of the cores are given in the separate supplement mentioned above.[16]

The coring data for the section at CRM 7.5 (Fig. 6) and for the other sections indicate that the radioactive sediments are thicker downstream from CRM 14, and this is generally confirmed by the sediment range surveys made periodically by TVA. The single core showing the greatest thickness of radioactive sediment, 8.7 ft, was obtained from Hole No. 5 at CRM 7.5. Upstream from CRM 14 radioactive sediments were largely on the sides of the stream channel. Downstream they were generally thickest on the inside of a bend and on the flatter side of the channel, but extended farther out into the main part of the stream.

From the data obtained by analyses of sediment cores in the downstream portions of the study reach it appeared that there was a recurrent general pattern of variation of gross gamma activity with depth of sediment. Also, the variations of sediment activity were very similar to variations in the annual quantities of ^{137}Cs released through White Oak Dam (see Figure 8). This similarity suggests some interesting implications. Radiological analyses of water samples from White Oak Creek at White Oak Dan and from Clinch River near Centers Ferry showed that cesium present in the water was almost wholly in the suspended sediments.[22] Therefore, it would be expected: (1) that ^{137}Cs would be incorporated into river bottom sediments primarily from sedimentation of suspended matter rather than from chemical interactions in the river; and (2) that, where sediment deposition was normal, variations of radiocesium with depth in the sediments should

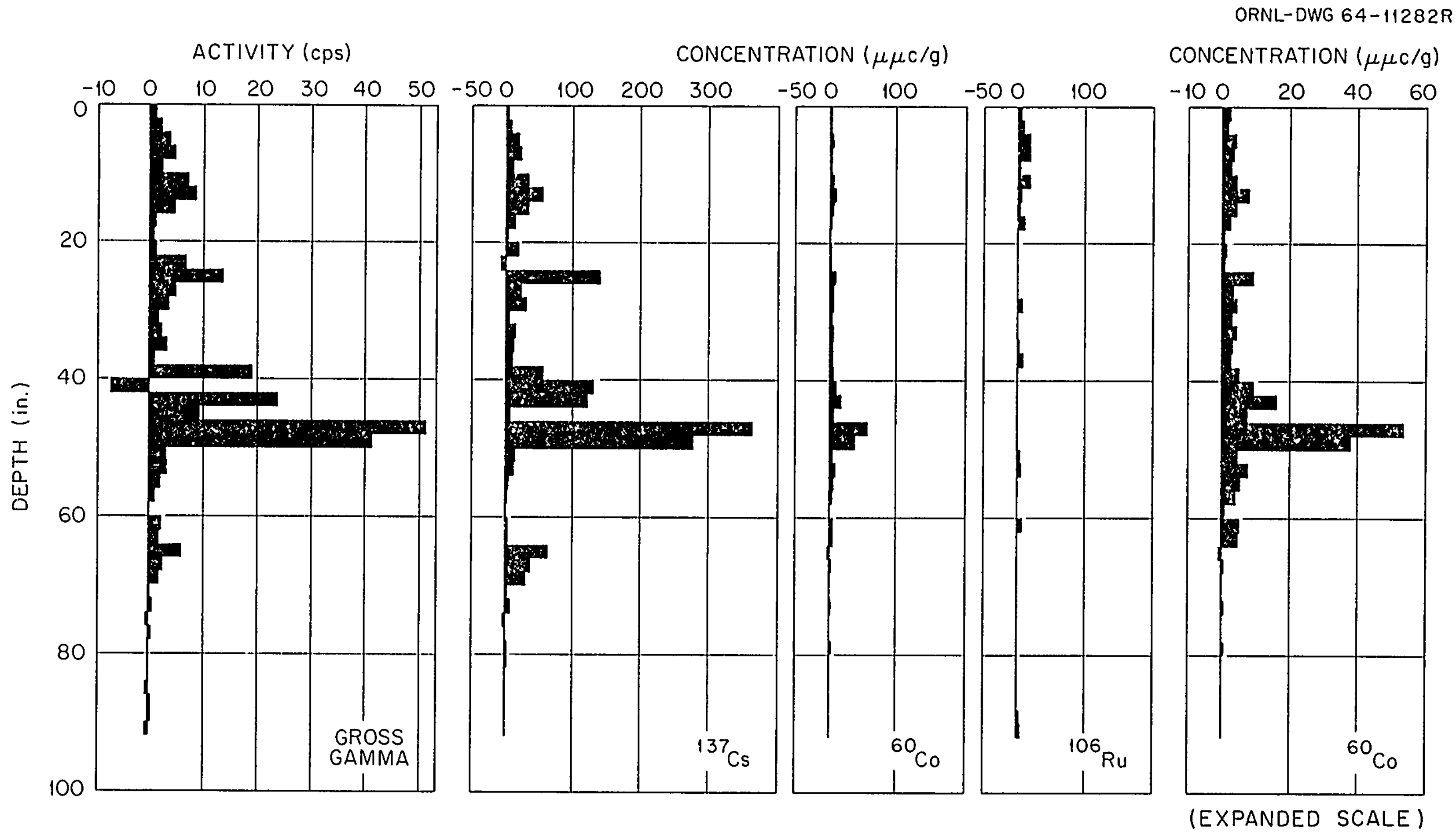

Fig. 8. Variations with Depth in Concentrations of Gross Gamma Activity, ^{137}Cs, ^{60}Co, and ^{106}Ru in Bottom Sediment Core from Hole 6, CRM 7.5.

reflect the variations in releases of ^{137}Cs in past years of which records are available for almost 20 years. In a general way the above appeared to be true.

Few of the cores appeared to show a pattern of gross gamma radioactivity such as would be expected if much of the relatively large releases of ^{106}Ru during the past few years had been incorporated into the river sediments (see Fig. 8). The relatively low levels of gross-gamma radioactivity in the upper layers of the cores suggested that a very small fraction of the ruthenium released to the river was in the sediments. This conclusion was supported by radionuclide mass-balance calculations based on analyses of 1961 Clinch River water samples.[8]

Geochemical Investigations

Because no standard methods have been devised for determining the chemical form of sediment constituents, it was thought advisable to begin work on the geochemical analyses of two selected cores obtained from widely separated river sections. After scanning for gross gamma counts of all cores and gamma spectrum counting of selected cores, the frozen cores were sliced longitudinally using a specially constructed power saw. The detailed work of analysis of the two selected cores was done at various laboratories of the USGS and at ORNL.

Each of the two cores was divided into a number of 2-inch increments on the basis of visual logging and radionuclide scans of each intact core. In addition to radionuclide analyses of the two cores various physical and chemical tests were made and desorption tests were performed. Also it was hoped that as a part of the analytical work the year in which specific core increments were deposited could be identified by comparing the core scans with the pattern of annual radionuclide releases through White Oak Dam. Dating of the cores in this way would provide information on the net sedimentation rates at the sampled cross sections. The extent to which these hopes were realized is indicated in the separate supplement.[16]

Behavior of Radionuclides Associated with Clinch River Sediments

Radionuclide desorption studies were made as a part of the Clinch River Study in order to determine the release characteristics of nuclides associated with sediments. This information is necessary in assessing the hazards of the systems. Desorption studies also are an aid in devising improved waste-disposal systems, and in defining the dominant natural forces responsible for the retention and migration of the radionuclides in the river system.

A sample of naturally contaminated bottom sediments was obtained from the mouth of White Oak Creek, CRM 20.8, where radioactivity was known to be relatively high. For laboratory studies to simulate naturally contaminated sediment, a bottom sediment sample was obtained from CRM 15.3, and standard clay minerals purchased from Ward's Natural Science Establishment were also used for some tests. Taking advantage of existing information about cesium and strontium sorption, simulated studies of these radionuclides were done with radioisotopes purchased from the ORNL Isotopes Division. Ruthenium studies were made with the ruthenium present in ORNL intermediate-activity waste (tank W-8), and the cobalt isotope was purchased from the Isotopes Division. Most of the work with the latter two elements involved characterization of their behavior in solutions.

Desorption of Radionuclides from Naturally Contaminated Sediment

Duplicate samples of the moist sediment from the mouth of White Oak Creek equivalent to 25 g of oven-dry weight were contacted with 200 ml of the various desorbing solutions. The pH was adjusted prior to contact, immediately after contact, and 4 hrs after contact. After standing for 24 hrs, the samples were centrifuged and the supernatant liquid was decanted. In order to raise the activity level for more accurate determination of the radionuclides, the supernatants from the duplicate samples were combined, acidified with HNO_3, and reduced in volume by heating prior to radiochemical analysis.

The results obtained are shown in Table 6. The radionuclide content of the sediment was (dis/min/50 g, oven-dry basis): ^{60}Co, 16,600; ^{137}Cs, 118,000; $^{103-106}Ru$, 129,000; ^{90}Sr, 2405; ^{144}Ce, 4145; and TRE, 24,190. Cesium-137 and ruthenium-103 and -106 were the principal contributors to the activity; and the distribution of the radionuclides in this sample agreed with the earlier results obtained with samples from the vicinity of CRM 20.8.

Up to 1 M solutions of salts, such as KCl, NaCl, $CaCl_2$ between pH 6 and 8, removed less than 10% of the ^{60}Co, ^{106}Ru, and ^{137}Cs; on the other hand, between 30 % and 77% of the ^{90}Sr was removed by the salts, depending on their concentration and pH. This behavior suggests that the strontium was held primarily by simple ion exchange, whereas cobalt, ruthenium, and cesium were not. In the strongly acid systems, as HNO_3 or HCl , cobalt and strontium were released, but ruthenium and cesium resisted desorption. In strongly alkaline systems approximately one-half of the ruthenium and approximately 15% of the cobalt were desorbed. Cesium and strontium were not desorbed at the high pH; and the 20% removal of strontium at pH 8 using NH_4OH was not confirmed by later tests.

Desorption of Cesium from Sediments

Intensive investigation of the behavior of cesium in waste solutions by the ORNL waste research group has shown that cesium sorption by sediments may be regarded as an

Table 6. Removal of Cobalt, Cesium, Ruthenium, and Strontium from Clinch River Sediment by Various Solutions

Reagent	Concentration (Molarity)	pH	% Removed			
			^{60}Co	^{137}Cs	^{106}Ru	^{90}Sr
Tap Water		6 (HNO_3)	2.8	---*	3.0	21.3
Tap Water		2 (HNO_3)	64.6	---	3.1	80.9
Tap Water		1 (HNO_3)	78.1	3.3	5.5	
Tap Water		6 (HCl)	---	---	3.5	19.4
Tap Water		2 (HCl)	65.6	---	4.1	89.9
Tap Water		7.7 (Natural)	---	---	4.5	11.0
$NaHSO_3$	0.1	6	16.8	---	9.8	37.7
$K_2Cr_2O_7$	0.1	5.6	5.9	1.7	4.7	73.1
$CaCl_2$	0.1	7	6.4	---	4.1	58.6
$CaCl_2$	1.0	7	4.7	---	4.4	76.9
NaCl	0.1	6	3.5	0.8	4.1	39.2
NaCl	0.1	8	---	5.7	6.8	30.1
NaCl	1.0	6	---	0.7	3.9	63.1
NaCl	1.0	8	---	0.6	5.1	56.0
KCl	0.1	6.2	---	0.7	2.8	53.0
KCl	1.0	6.2	3.0	1.7	2.5	68.7
NaOH		8	---	0.4	5.9	6.0
NaOH		12	16.5	3.1	46.6	4.9
NH_4OH		8	---	0.7	8.2	20.0
NH_4OH		11.8	17.2	4.5	45.1	3.5
Ethyl Alcohol					0.9	<1
Acetone					1.0	

*--- Refers To Concentrations Below Detectable Limits.

ion exchange reaction complicated by exchange sites which have an extremely high affinity for cesium. These sites for cesium are present because of the layered arrangement in minerals which, in illite, provides a favorable spacing for retention of cesium ions (c-spacing $\cong 10$ Å). Desorption studies, reported elsewhere, show that minerals such as illite or muscovite can even resist the attack of acids and retain the cesium.[23]

With the mineral illite, potassium at the edges of the crystallite can be exchanged for cesium. Actually less than 1% of the total potassium in the mineral illite is exchangeable and this comprises 10% of the total exchange capacity. Once bonded in the interlayer site near the edges, cesium is difficult to replace or remove and with longer contact time the cesium diffuses further into the lattice of the mineral and removal becomes even more difficult. It is believed that the cesium on the sediment is associated with illite or mica in this manner.

If cesium or strontium is held in readily accessible sites, such as those found on montmorillonites or kaolinites, desorption can be accomplished by contacting with high concentrations of salt solutions. Hence, even with illitic material, ion exchange reactions occur at or near the surface, and ions held at these sites are readily exchanged. It is believed that strontium is held primarily by this mechanism.

Desorption of Strontium from Sediments

The data in Table 7 show that tap water at pH 2 removed more strontium than 1 M $CaCl_2$ or NaCl. Simulated tests using sediment from CRM 15.3 suggested that strontium which sorbed at pH 8 should be desorbable equally or perhaps more efficiently by $CaCl_2$ than by tap water adjusted to pH 2. At pH 8 the sample from CRM 15.3 sorbed approximately 46% of the strontium (Table 7). This gave a

Table 7. Equilibrium Sorption and 45 hr Desorption of Strontium Using 1.0 Gram of CRM 15.3 Sediment in 100 ml of Tap Water at Indicated pH

Sorption pH	8	10.8	10.2*	10.1**
Sorption (%)	46.2	93.2	94.1	68.4
Desorption (%)				
Tap H_2O; pH 2 (HNO_3)	96.3	95.3	100	89.3
Tap H_2O; pH 12 (NaOH)	14.7	5.1	3.6	
1 *M* $CaCl_2$ pH 7	97.8	68.8	61.2	5.8
1 *M* NaCl pH 8	90.2	83.0	100	85.3

*95 ml Tap Water + 5 ml Simulated Waste + Sr-85;
**95 ml Tap Water + 5 ml Simulated Waste + Sr-85; (No Sediment Added).

K_d of 85 ml/g which is a reasonable approximation of the K_d of 65 ml/g for ^{90}Sr on the naturally contaminated sediment.

On the other hand, as shown in Table 7, at pH 10.8 a much larger percentage of the strontium was sorbed (93.2%), and a K_d of 1300 ml/g was calculated. Using the desorption data in Table 6 for either NH_4OH or NaOH at pH 12, K_d's of 200 and 155 ml/g were obtained. These differences suggest that another mechanism, in addition to clay exchange, was operative when sorption occurred at high pH. It became apparent that $CaCO_3$ was being formed and the higher removal of strontium at pH 10.8 could be likened to the lime-soda softening process whereby strontium is removed from the solution mainly by the formation of $CaCO_3$.

Data on the sorption of strontium simply by diluting simulated waste with tap water without sediment is also shown in Table 7. The pH was 10.1; and the removal of strontium by the fine colloidal precipitate averaged 68.4%. The desorption data also show that $CaCl_2$ was ineffective as a desorbing agent for this precipitate. The explanation given for this was that when sorption of strontium by sediments was made to take place at pH 10, strontium was sorbed not only by the clay by ion exchange but on the precipitate of calcium carbonate. The $CaCO_3$ was formed as the carbonate ion from the bicarbonate ion in tap water. When the $CaCl_2$ solution at neutral pH was added, the strontium on the ion-exchange mineral complex was displaced but the strontium on the precipitate was not.

Studies are continuing on the role of $CaCO_3$ formation on strontium sorption. Tests with a sample of sediment from White Oak Lake suggest that $CaCO_3$ may play an important role in retaining strontium. With 1 M $CaCl_2$ only 35% of the strontium was removed, compared to 77% removal from the sediment obtained at CRM 20.8 (Table 6). The lower removal from the White Oak Lake sediment suggests that an even greater percentage of the strontium was associated with $CaCO_3$. Further evidence to support the important role of $CaCO_3$ was found through analysis of the sediment; the sediment from White Oak Lake contained 4.22% $CaCO_3$ while the sediment from CRM 20.8 contained only 1.77%.

Desorption of Cobalt from Sediments

Although understanding of the mode of occurrence of cobalt and ruthenium is limited, certain responses of these nuclides in demineralized and tap water, and with sediment as well as resin exchanger, are given below.

When a ^{60}Co spike (approximately 10^{-7} $\underline{M}$) was mixed with demineralized water or tap water adjusted to pH 2 and the pH of the mixture was gradually increased without any sediment present, the percentage removal of cobalt increased in both the demineralized and tap water systems until a pH of 9 was reached. Above this pH cobalt removal decreased slightly in the demineralized water system, but increased sharply in the tap water system (see Fig. 9). The high removal of cobalt in the tap water system was caused by co-precipitation of cobalt with calcium carbonate above pH 9. With the concentration of cobalt added to the demineralized water, no precipitate of cobalt hydroxide would be expected until the pH is at least above 9.5. A possible explanation of the removal of cobalt from demineralized water is adsorption on the glass sides of the container.

In the sorption of cobalt from demineralized water by sediments optimum removal was obtained between pH 6 and 8, similar to the behavior of cobalt in demineralized water without sediment. On the other hand with Dowex 50, maximum sorption occurred at pH 4 and the higher pH levels showed a gradual reduction in sorption. On desorbing the cobalt, the Dowex 50 released all of the cobalt with neutral sodium acetate; but only about 80% of the cobalt was removed from the sediment with the same treatment.

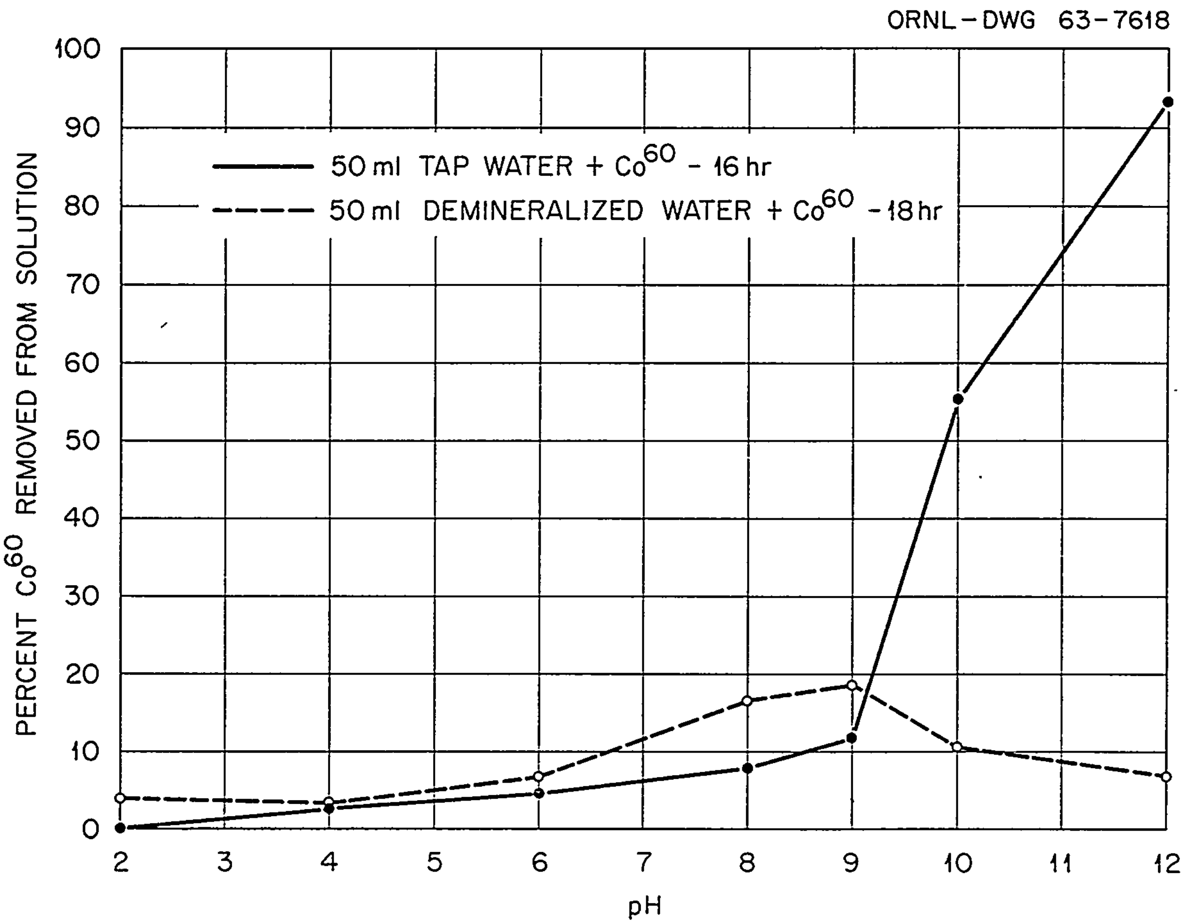

Fig. 9. Removal of Cobalt from pH-Adjusted Tap Water and Demineralized Water as a Function of pH

Since neutral salts did not remove cobalt from the naturally contaminated sediment to any appreciable degree, simple ion exchange was eliminated from consideration. Two possible factors which must be considered in more detail to gain further insight into the mechanism include: (1) an understanding of the surface features of clays and sediments, and (2) more extensive investigation of the properties of the cobalt ion at different pH levels. The latter includes the possibility of forming monohydroxy cobalt prior to the formation of the dihydroxy form.

Desorption of Ruthenium from Sediments

Alkaline pH conditions were found to be more effective for desorbing ruthenium from the sediment. Since early studies on ruthenium from waste seepage systems showed that the ruthenium is not present as $Ru(NO_3)_3$ or $RuCl_3$ salts, tests in the laboratory to simulate natural contamination were performed using ruthenium from the ORNL waste tanks. The waste contained (dis/min/ml) ^{106}Ru, 1.84×10^6; ^{137}Cs, 1.44×10^6; ^{90}Sr, 6.34×10^3 as principal radionuclides. Paper partition chromatography methods for the identification of ruthenium complexes indicated that the ruthenium in the waste was either nitrosyl ruthenium hydroxide $\left[(RuNO(OH)_3(H_20)_2)\right]$ or mono nitrato nitrosyl ruthenium hydroxide $\left[(RuNO(NO_3)(OH)_2(H_20)_2)\right]$. The mono nitrato nitrosyl ruthenium is reported by Story and Gloyna[24] to be cationic and exchangeable; the low desorption of ruthenium by the sediment with neutral salts suggests that the ruthenium is not easily exchangeable and further suggests that the sorbed ruthenium may be in the nitrosyl ruthenium hydroxide form.

To compare the desorption properties of ruthenium sorbed on a sediment using the waste solution, a sample of sediment from CRM 15.3 was contacted (for sorption) with a solution containing 2 ml of waste and 98 ml of tap water adjusted to pH 7. After 24-hr contact approximately 58% of the ruthenium was removed from the solution by sorption (Table 8). The samples were then leached with the solutions listed in Table 8. The desorptive behavior of the sediment was similar to that observed with the naturally contaminated sediment; however, desorption from the artificially contaminated sediment was greater than from the naturally contaminated sediment.

Observations of the waste on dilution with tap water offered a possible explanation of the higher desorption with the artificially contaminated sediment. With 2 ml of waste and 98 ml of tap water adjusted to pH 7, a colloidal precipitate was observed in the system; and centrifugation showed that approximately 20% of the ruthenium was associated with this colloidal precipitate. From analysis of the waste it was presumed that the precipitate was aluminum oxide hydrate; considerations of the solubility product precluded the formation of calcium carbonate at this pH.

When the pH of the solution containing the precipitate was lowered to pH 2, 50% of the ruthenium returned to the solution and the precipitate dissolved. If one assumes that 34% (20% ÷ 58%) of the ruthenium in the sediment is associated with the precipitate which releases one-half of its ruthenium as it dissolves at pH 2, then of the amount desorbed at pH 2, 17% is due to the precipitate and about 6% is due to desorption from the sediment. The latter value agrees well with desorption of ruthenium from naturally contaminated sediment. Further speculation is premature at this time.

Table 8. Sorption and Desorption of Ruthenium using 10.0 g of CRM 15.3 Sediment and 100 ml Solution

Test Number:	1	2	3
SORPTION:			
SOLUTION	(98 ml TAP WATER + 2 ml WASTE FROM TANK W-8)		
pH	7	7	7
4 Hr (%)	52.5	56.0	54.1
24 Hr (%)	58.8	60.1	55.3
DESORPTION:			
SOLUTION	TAP WATER	1 *M* $CaCl_2$	TAP WATER
pH (Adjusted with HNO_3 or NaOH)	2 (Adjusted with HNO_3 or NaOH)	7	12 (Adjusted with HNO_3 or NaOH)
4 Hr (%)	27.5	9.5	53.8
72 Hr (%)	23.3	12.2	61.8

Longitudinal Distribution of Radionuclides in River Sediments

Relationships in Distribution in Bottom Sediments of the Clinch and Tennessee Rivers

Similarities in the longitudinal distribution of radionuclides in the bottom sediments of the Clinch and Tennessee Rivers were studied using data from the 1961 annual survey by the ORNL Applied Health Physics Section. This sediment survey in 1961 extended from near the mouth of White Oak Creek on the Clinch River to near Kentucky Dam on the Tennessee River. Using methods described by Cottrell,[17] sediment samples were collected at 32 sections; and the concentrations of ^{137}Cs, ^{60}Co, ^{106}Ru, ^{144}Ce, other rare earths, ^{95}Zr- ^{95}Nb, and ^{90}Sr in a composite sample of the bottom sediments at each section were determined.

Preliminary graphical comparison of the concentrations of ^{137}Cs and ^{60}Co at each section indicated a straight-line relationship on log-log paper. To better measure the relationship, the index of correlation, the standard error of estimate, and the regression coefficient were computed for comparison of the logarithms of the concentrations of these two radionuclides. Similar computations were made for all pairings of the logarithms of the concentrations of radionuclides using a computer program, adapted from a general and basic correlation program prepared by A. M. Craig, Jr., of the ORNL Mathematics Division.

The results of some of these computations are shown in Fig. 10 in which the concentration of ^{137}Cs is treated as the "independent" variable. A tabulation of the several indices of correlation showed that the choice of other radionuclides for use as the independent variable in Fig. 10 would have been appropriate. It should be noted that determination of the concentration of rare earths includes ^{90}Y but excludes ^{144}Ce.

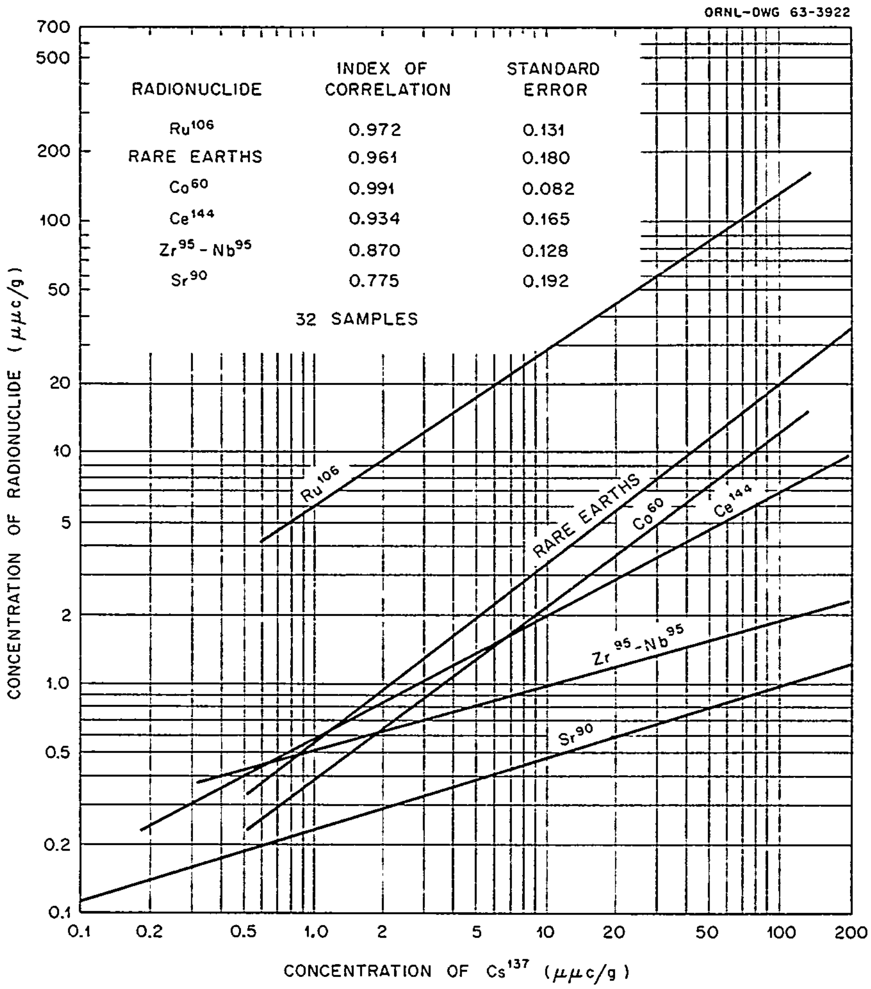

Fig. 10. Correlations Showing Similarity in the Longitudinal Distribution of Radionuclides in the Bottom Sediments of the Clinch and Tennessee Rivers

The slope of the curves is nearly the same for the logarithmic correlation of ^{137}Cs to ^{106}Ru, to the trivalent rare earths, and to ^{60}Co; and the indices of correlation for these curves are good. Thus the results shown in Fig. 10 suggest that similar mechanisms control the longitudinal distribution of these particular radionuclides.

The observed rates of decrease in concentration of cesium, ruthenium, cobalt, and the rare earths with distance downstream from White Oak Creek are much greater than rates calculated on the basis of flow dilution alone. This lack of relationship between concentration and flow dilution and the similar longitudinal distribution pattern of the radionuclides is a further indication that the mechanism controlling the distribution pattern is sedimentation rather than the chemical process of equilibration (reversible sorption) between the radionuclide concentrations in the sediments and in the water. Furthermore, work on the behavior of radionuclides associated with the sediments (pp. 52 to 62) has indicated that once the radionuclides are incorporated into the bottom sediments the potential for release of this radioactivity through desorption is negligible.

The slope of the curves of ^{90}Zr – ^{95}Nb and for ^{90}Sr in Fig. 10 is less than that for cobalt, ruthenium, or the rare earths, indicating that different mechanisms may be controlling the distribution of zirconsium-niobium and strontium in the bottom sediments.

The good correlations indicate that the longitudinal distribution of several radionuclides in the bottom sediment may be determined on the basis of the distribution of one or two of these radionuclides.

Although not specifically cited earlier in this report (pp. 34–35) the relative longitudinal changes in ^{90}Sr concentration in the bottom sediments have been found to be quite similar to changes that would have been attributed to the effects of flow dilution. It is quite probable that the most important mechanism controlling the longitudinal distribution of ^{90}Sr is flow dilution.

Seasonal Changes in Distribution of Radioactivity in Bottom Sediments of the Clinch River

In general, the over-all longitudinal trend in levels of radiation in bottom sediments of the Clinch River is for the levels to increase as the distance downstream from White Oak Creek increases (see Fig.11). This tendency has been ascribed to hydraulic factors that may influence sediment deposition: larger flow areas, lower velocities, and greater surface area. However, the change in radiation levels with distance is not regular. In particular relatively high levels of radiation have been observed, in summer surveys, at several sections downstream from CRM 12.

Possibly the higher zones of radiation levels downstream from CRM 12 are the result of seasonal variations in these and other hydraulic factors; in the winter season greater discharges and increased velocities of flow occur; in the summer thermal stratification occurs under certain conditions.

From early fall to late spring the minimal water levels in the Clinch River below CRM 20.8 are about 6 feet lower than in the remaining portion of the year. On several days during this winter period, discharges may exceed 15,000-20,000 cfs. During the summer period, from late spring to early fall, the discharge has rarely exceeded 10,000 cfs. Because of the greater discharges and lower water levels the velocities in the winter period are greater than in the summer throughout the study reach.

Cold waters released from Norris Reservoir in the summer period cause the flow in the study reach to be thermally stratified and these cold waters underflow the

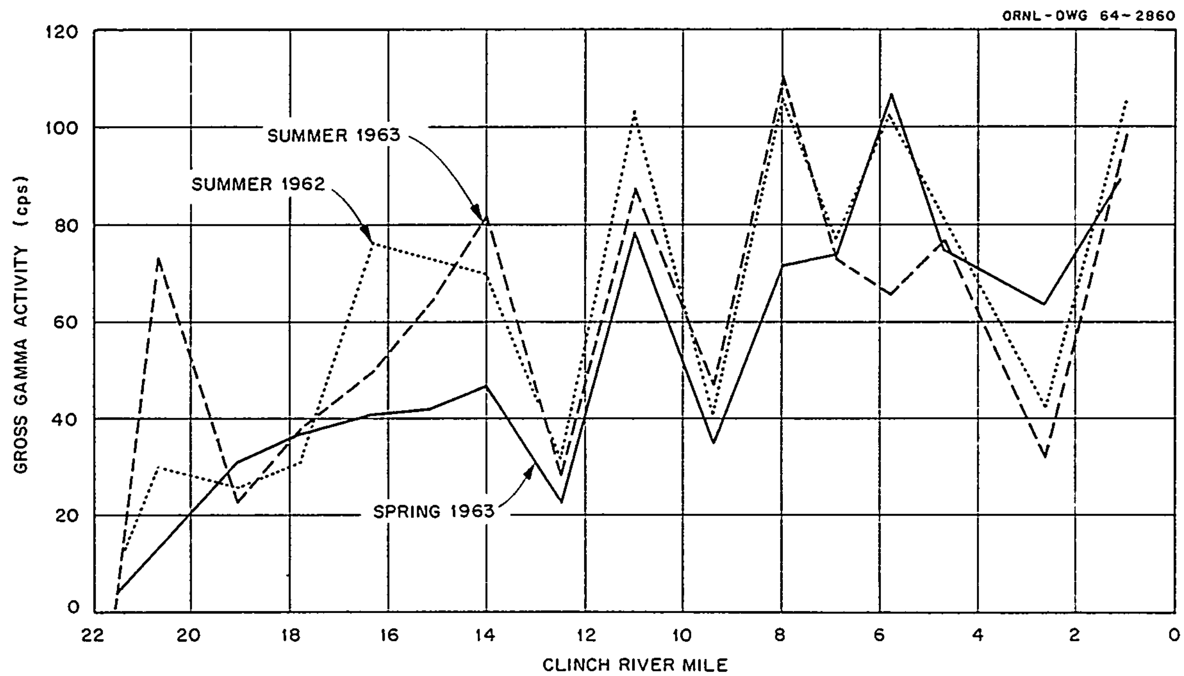

Fig. 11. Longitudinal Variation in Average Sectional Gamma Count Rate at the Surface of Bottom Sediments in Clinch River.

essentially stagnant warm water backed up from the main body of Watts Bar Reservoir. This condition which tends to restrict the effective flow area in the river is shown schematically in Fig. 12. On the basis of median discharges for the summer months during the period 1947-61 it may be estimated that the duck under centers around CRM 9.5 (see Fig. 13).

The influence of the seasonal changes in the various hydraulic factors was investigated by a survey of gamma radiation at the surface of bottom sediment by the ORNL Waste Disposal Research Section and USGS in the spring of 1963 (April 30 to May 8). This spring survey (1963) was made as soon after the winter high flows as practicable using the method reported by Cottrell.[17]

In Fig. 11 the variations in average gamma count rate at the surface of the sediment at the various cross-sections in the Clinch River is shown for three surveys: summer surveys of 1962 and 1963 and spring survey of 1963. The results of the surveys in Fig. 11 do not indicate that the decrease in effective flow area due to thermal stratification in the summer period causes any major change in the longitudinal variation of radioactivity in the bottom sediments. If the relative pattern of longitudinal variation in gamma count rates had been influenced by thermal stratification the general longitudinal trend toward increasing radiation levels in the downstream direction for summer conditions would have been reversed in the vicinity of CRM 9.5.

It can only be considered as being somewhat fortuitous that the absolute variations as well as the relative variations in gamma count rates shown in Fig. 11 are about the same for all curves. Study of data for the releases of radioactivity through White Oak Dam indicate that this is because the loads of radioactivity released were generally on the decline throughout the period preceding the three surveys.

ORNL-DWG 64-2861

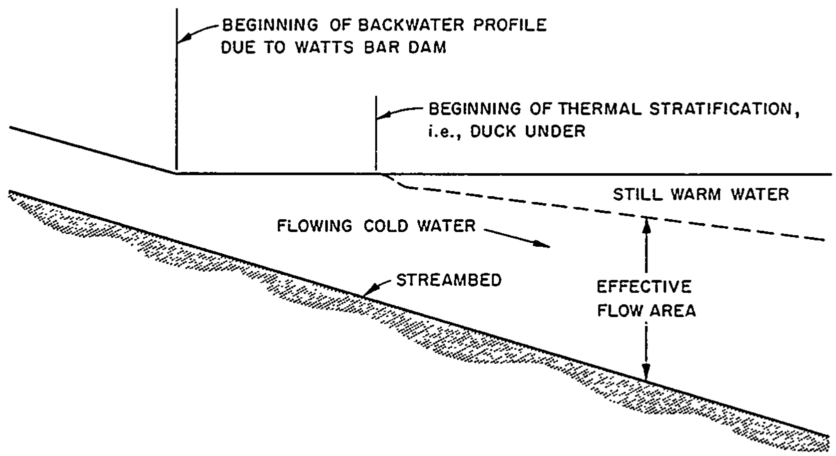

Fig. 12. Schematic Sketch Showing Effect of Thermal Stratification on Effective Flow Area in Clinch River

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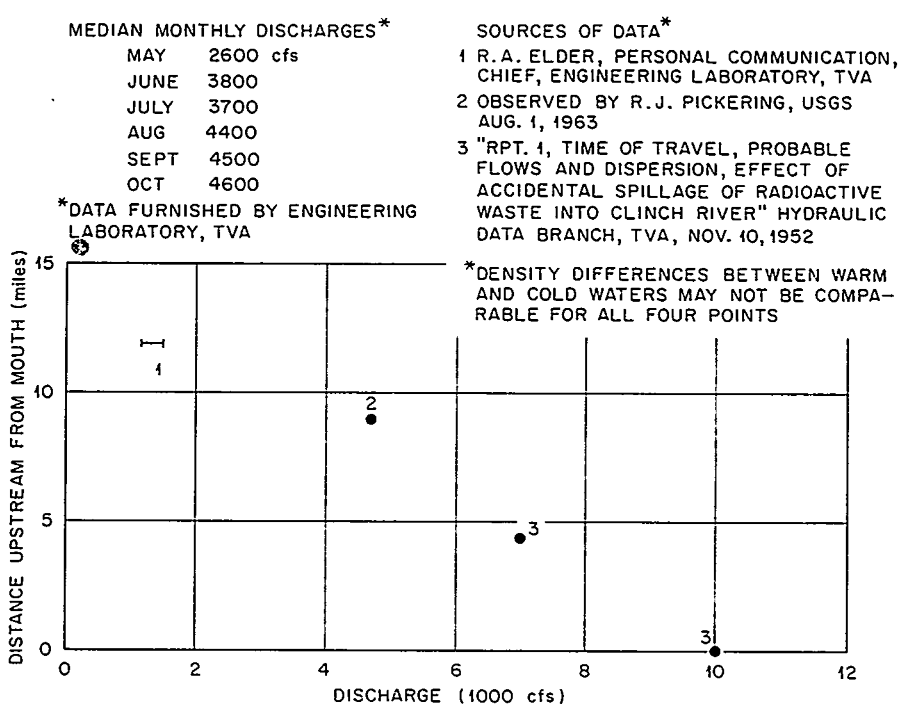


Fig. 13. Approximate Relation for Location of Duck Under

Survey of Bottom-Sediment Radioactivity in Clinch River Sloughs

During May, 1963 a survey of bottom-sediment radioactivity was made in six large sloughs (drowned tributary stream mouths) of the Clinch River and one slough of the Emory River (see Fig. 14). The primary purpose of the survey was to determine whether the sloughs represent repositories of fine sediment with higher surface radioactivity than that in the main channel of the river. Radioactivity readings were made with the Flounder instrument, obtained from the ORNL Applied Health Physics Section. The results of the survey are shown in Table 9.

The results indicate that the radioactivity of sediment in the sloughs surveyed is within the range of radioactivity in the main river channel and in some places is several times the "background" (surface soil) radioactivity. However, in none of the sloughs surveyed was the maximum radioactivity as high as the maximum radioactivity in adjacent portions of the main channel.

Plots of bottom sediment radioactivity versus depth of water indicate that in several of the deeper sloughs, there is a direct relationship between those two variables. Although the causes of this relationship are not definitely known, it is probable that two erosional processes are largely responsible: (1) exposure of the portions of the sloughs less than six feet deep to subaerial erosion during the winter period of low water in Watts Bar Reservoir, and (2) washing of sediment down the sides and into the deeper portions of the sloughs during periods of high rainfall and high surface runoff.

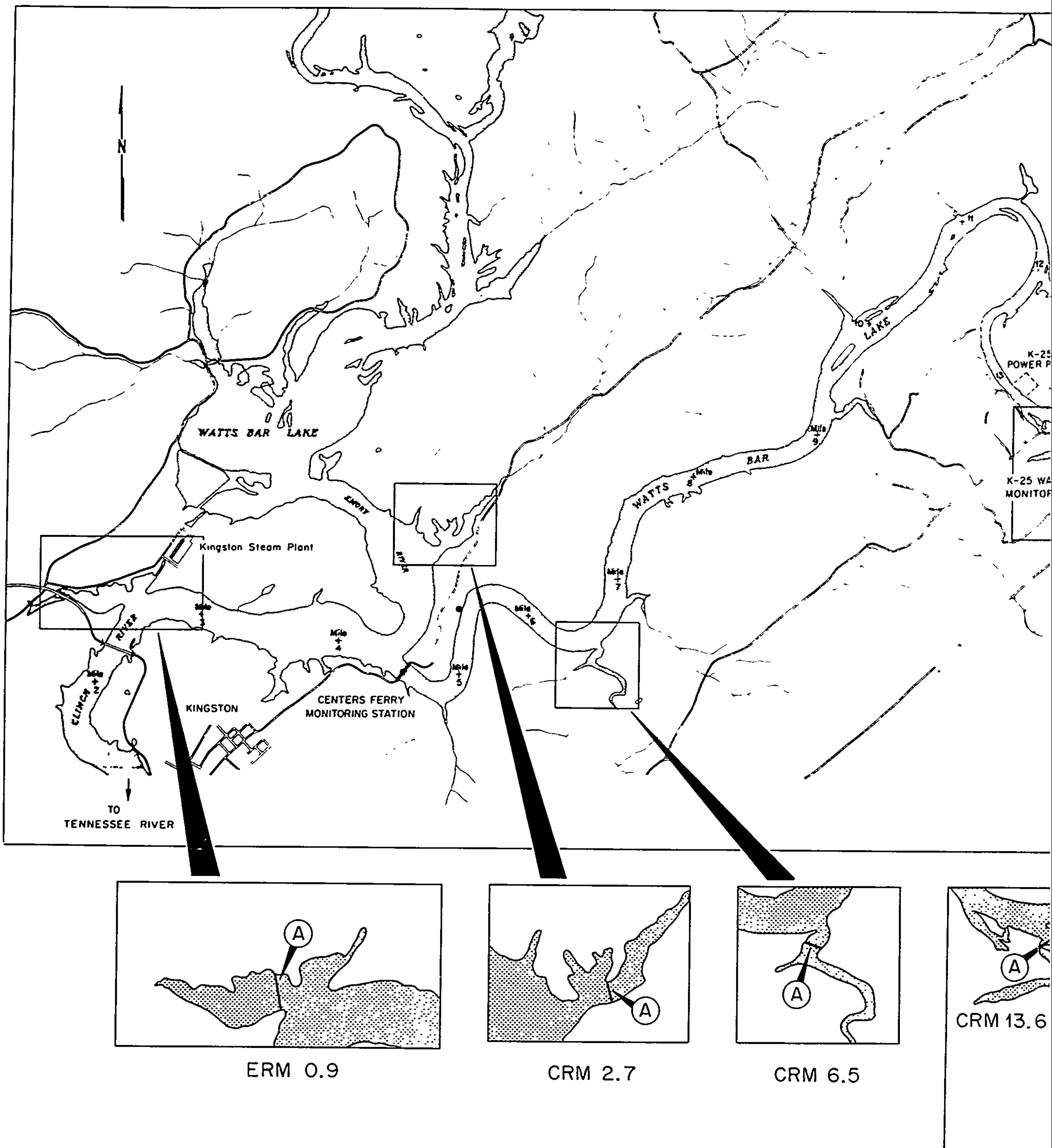
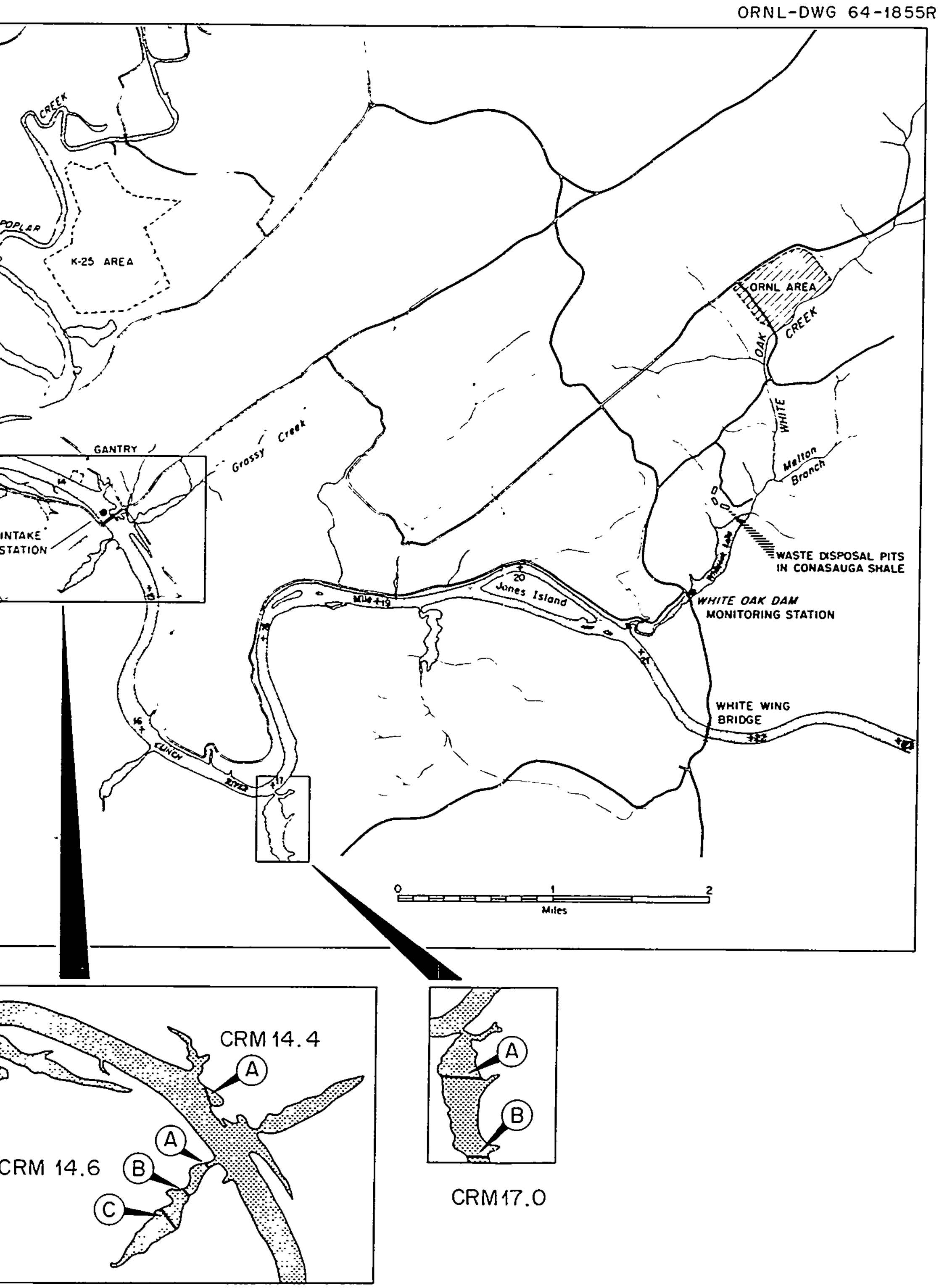


Fig. 14. Location Map, Survey of Bottom
in Clinch River Sloughs



diment Radioactivity

Table 9. Bottom Sediment Radioactivity in Sloughs

			Radioactivity cps			
River	Mile*	Section	Average	Maximum	Minimum	Background**
Clinch	17.0	A	13.0	17.2	9.7	7.8
		B	7.9	9.4	5.1	N.D.
	14.6	A	16.0	42.0	7.9	6.5
		B	15.2	26.9	6.9	6.6
		C	15.3	20.8	10.6	N.D.
	14.4	A	28.6	54.4	11.1	N.D.
	13.6	A	34.5	53.3	11.1	6.5
	6.5	A	21.3	32.8	4.1	6.2
	2.7	A	19.8	38.1	5.8	N.D.
Emory	0.9	A	15.0	20.8	8.2	7.2

N.D. - Not determined.

* Miles upstream from mouth of the river.

** Background determined on surface of soil at bank of slough.

Sediment Survey of the Clinch and Tennessee Rivers

Annual surveys of sediments in the Clinch and Tennessee Rivers were made by the ORNL Applied Health Physics Section during the summers of 1962 and 1963. In both years, the survey of the Tennessee River extended from Fort Loudoun Reservoir (TRM 615.8) into Guntersville Reservoir (TRM 354.4). The 1963 survey of the Clinch River extended upstream from the mouth of the river to CRM 42.8 (~ 20 miles above Melton Hill Dam). The techniques and procedures used were those described in 1959 by Cottrell.[17]

Figures 15 and 16 show the gamma count rate at the surface of the Clinch and Tennessee River bottom silt according to river miles for the years 1961, 1962 and 1963. The longitudinal dispersion of gamma emitting radioactive materials in the silt of the Clinch River in 1962 (Fig. 15) was essentially the same as that in 1961 but the levels of radioactivity measured were lower. The lower levels detected correlate with the decrease in the total amount of radioactivity released to the river in 1962. A total of 2187 curies was discharged to the Clinch River during the 12-month period just prior to the 1961 survey, while only 1700 curies were released during the corresponding period in 1962.

The increase in gamma count rate of the Tennessee River silt in 1962 (Fig. 16) appears to be due primarily to fallout from weapons testing. This conclusion is supported by the fact that background readings in the Tennessee River silt (Fig. 16) and Fort Loudoun Reservoir silt (not shown) were higher in 1962. Fort Loudoun background readings increased from 8.9 c/s in 1961 to 16 c/s in 1962.

Table 10 shows the average concentrations of the major radionuclides found in Clinch River water, upstream from White Oak Creek, at the mouth of White Oak Creek, and downstream from White Oak Creek, respectively. The data in Table 10 (A) also support the conclusion that the 1962 river survey data were influenced by fallout. Increased average concentrations of ^{90}Sr, $^{103-106}Ru$, ^{95}Zr–^{95}Nb, and ^{144}Ce were detected in the Clinch River (CRM 41.5) upstream from the mouth of White Oak Creek. Such increases can only be attributed to weapons fallout. The increased concentrations of fallout material in the river water would have very little effect upon the relatively high gamma count rate in the Clinch River but would influence significantly the relatively low count rate of the Tennessee River silt.

The 1963 survey showed the dispersal pattern of radioactive silt in the Clinch River to be essentially the same as in 1961 and 1962 (Fig. 15) except for the stretch of river immediately downstream from the mouth of White Oak Creek and immediately upstream from the point of entry of the Emory River. The average of all gamma measurements taken in the Clinch River, downstream from CRM 21.5, was approximately the same in 1963 (47 c/s) as in 1962 (49 c/s). The total number of curies discharged to the river, however, decreased from 1700 for the 12-month period just prior to the 1962 survey to 794 during the corresponding period in 1963.

When compared to the 1962 data, the gamma count rate on the bottom silt of the Tennessee River in 1963 showed decreases in Watts Bar and Hales Bar Reservoirs and essentially no change in Chickamauga and Guntersville Reservoirs (Fig. 16).

Table 10. Average Concentrations of Major Radionuclides in Clinch River Water at Three Stations, 1961 and 1962.

(Units of 10^{-8} μc/ml)

Period	Radionuclide					
	^{90}Sr	^{144}Ce	^{137}Cs	$^{103-106}Ru$	^{60}Co	$^{95}Zr-Nb$
A. UPSTREAM FROM WHITE OAK CREEK, CRM 41.5[a]						
Third Qtr.,1961	0.10	0.05	*	0.45	*	*
Fourth Qtr.,1961	0.08	0.04	0.05	0.59	0.06	0.32
First Half, 1962	0.20	0.17	0.01	0.90	*	0.68
B. AT MOUTH OF WHITE OAK CREEK, CRM 20.8[b]						
Third Qtr., 1961	0.23	0.02	0.17	9.0	0.13	3.1
Fourth Qtr.,1961	0.28	0.10	0.25	37	0.30	4.9
First Half, 1962	0.22	0.03	0.12	18	0.24	0.24
C. AT CENTERS FERRY, CRM 4.5						
Third Qtr, 1961	0.25	0.11	0.05	5.9	0.02	*
Fourth Qtr., 1961	0.33	0.18	0.09	21	0.36	0.90
First Half, 1962	0.41	0.27	0.12	21	0.41	0.74

[a]Sampling station moved from CRM 33.2 to CRM 41.5 about January 1, 1962.

[b]Values given for this location are calculated concentrations resulting from ORNL waste releases based on concentrations of radionuclides in White Oak Creek and the dilution afforded by the river; they do not include amounts of radioactive materials (e.g., fallout) that may have entered the river upstream from CRM 20.8.

*None detected.

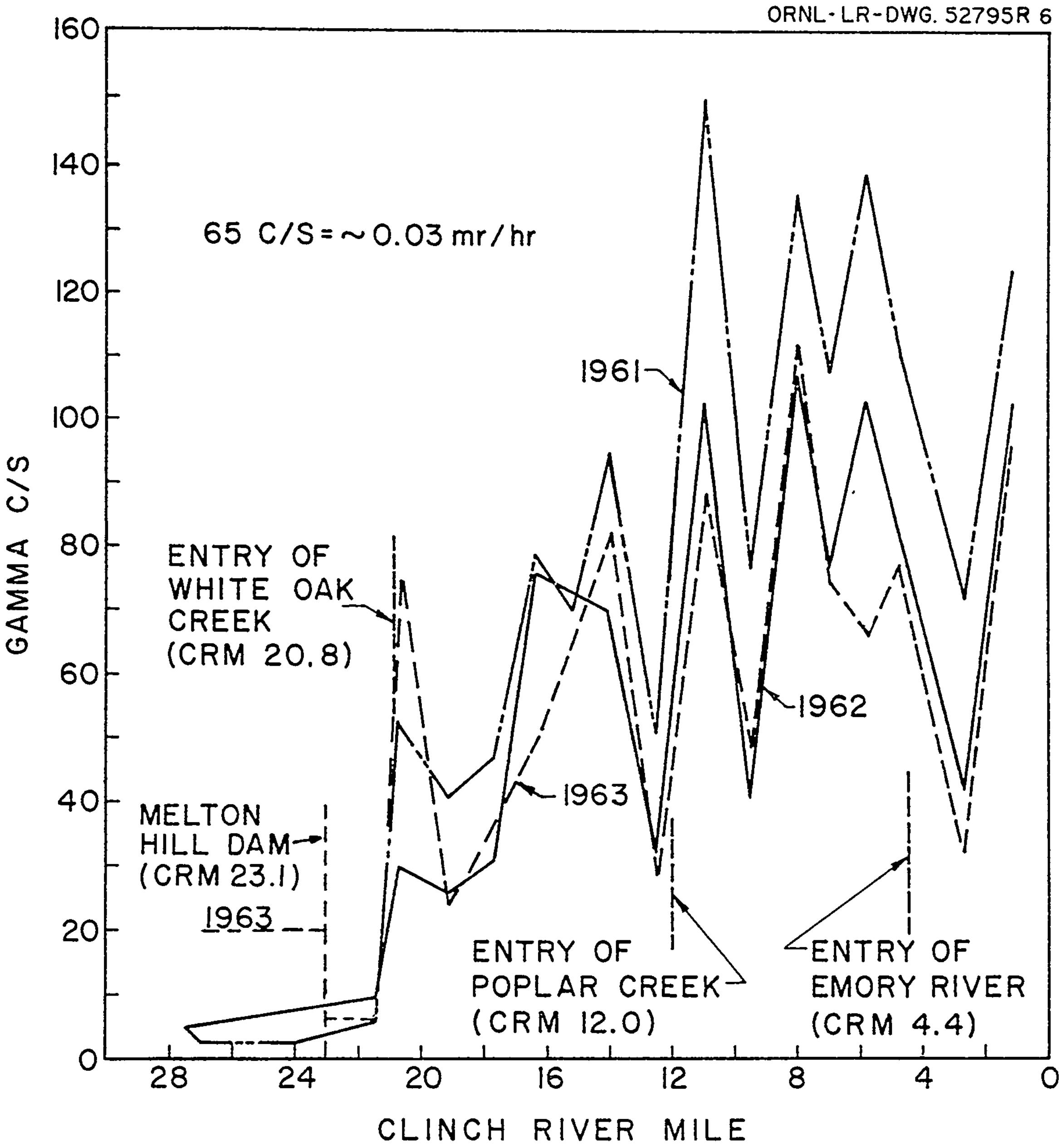


Fig. 15. Gamma Count at Surface of Clinch River Silt

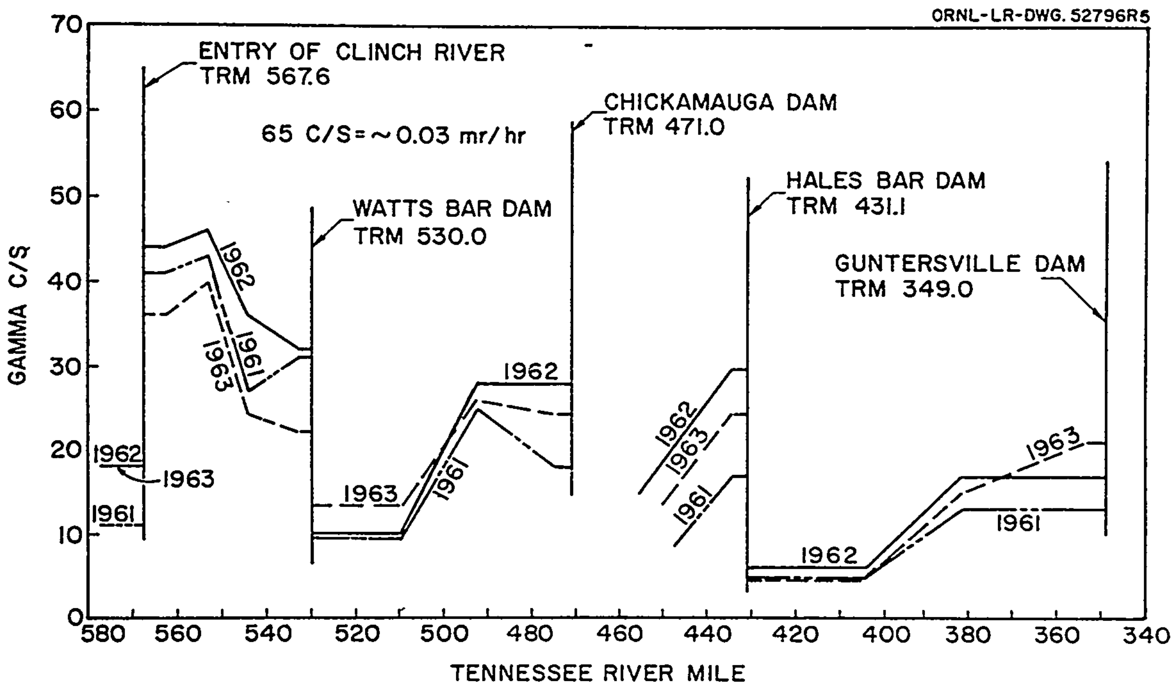


Fig. 16. Gamma Count at Surface of Tennessee River Silt

A comparison of the number of curies of each radionuclide discharged to the Clinch River with the average concentrations (μ c/g) of these radionuclides found in samples of the Clinch and Tennessee River silt during the period 1954-1961 is given in Status Report No. 3, Fig. 13.[3] The data on radionuclides discharged represent the number of curies of each radionuclide discharged during the year (July-June, inclusive) prior to each survey.

Tables 11 and 12 respectively, show the concentrations of radionuclides found in the Clinch and Tennessee River silt at various river-mile locations for the years 1962 and 1963. Fort Loudoun Reservoir background data are given for comparative purposes in Tables 12.

Radioactivity in Dredged Material Removed from Clinch River During Channel Improvement*

In connection with the Melton Hill Project of the Tennessee Valley Authority (TVA), a navigation lock was constructed at the dam. To facilitate navigational use, channel improvement below the dam was required in three reaches (see Fig. 17): (1) CRM 18.3 to CRM 18.8 --adjacent to Grubb Islands; (2) CRM 19.5 to CRM 20.8--adjacent to Jones Island (sometimes called Blue Springs Island); (3) CRM 22.4 to CRM 23.0--immediately below Melton Hill Dam (CRM 23.1). Radiological hazards that might be encountered in the dredging and disposal of presumably contaminated sediments were investigated. The results of this investigation were reported to the Steering Committee on December 4, 1963.[6]

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*Based on a report by O. W. Kochtitzky, Chief, Radiological Health Staff, Division of Health and Safety, Tennessee Valley Authority at open meeting of the Steering Committee, December 4, 1963.[6]

Table 11. Radionuclides in Clinch River Silt – 1962–1963

(Units of 10^{-6} μc/g of Dried Mud)

Location	^{137}Cs		^{144}Ce		^{90}Sr		^{60}Co		$^{103-106}Ru$		$^{95}Zr-^{95}Nb$		TRE + ^{90}Y (as ^{90}Y)	
	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963
CRM 42.8		2.6		17		0.29		*		14		14		15
39.1		1.3		7.7		0.49		*		6.4		6.3		6.3
34.7		2.6		19		0.34		*		14		16		16
31.1		2.7		16		0.36		*		13		12		9
29.0		2.7		16		0.52		*		12		13		9
27.0		3.0		22		0.50		*		16		19		15
24.9		3.0		20		0.52		*		15		17		14
23.4		0.90		3.8		0.43		*		2.8		3.4		3.3
Average		2.4		14		0.43				12		13		11
CRM 21.5	3.2	2.7	11	0.43	0.36	0.63	*	*	11	0.45	16	0.14	10	0.89
19.1	5.2	2.9	3.8	0.90	0.41	0.74	0.72	1.9	6.1	4.4	6.2	0.90	3.5	5.2
16.3	58	218	5.2	4.2	0.72	3.1	8.1	16	50	17	3.9	2.2	14	34
15.2	55	16	5.2	3.5	0.90	0.81	7.3	2.8	46	13	4.2	3.8	13	5.6
14.0	237	150	6.2	5.0	1.8	1.7	20	12	43	29	3.6	3.4	31	23
11.0	63	75	6.9	8.8	1.0	1.2	8.6	8.0	68	35	5.4	7.4	16	22
8.0	59	62	8.5	8.8	1.0	0.90	8.6	8.9	70	48	5.4	6.9	18	16
5.8	94	67	8.4	12	1.6	1.3	12	9.5	68	49	6.5	10	22	19
4.7	86	53	9.5	11	1.2	1.4	14	8.5	86	44	6.0	9.7	22	17
2.6	73	63	7.7	6.7	0.72	0.81	10	7.7	77	26	5.6	5.4	16	17
1.1	56	68	13	17	0.72	1.4	9.0	8.5	76	50	11	17	16	23
Average	72	71	7.7	7.1	0.95	1.3	9.8	8.4	55	55	6.7	6.1	17	17

[a]Total Rare Earths minus cerium

*None detected

Table 12. Radionuclides in Tennessee River Silt - 1962-1963

(Units of 10^{-6} μc/g of Dried Mud)

Location		^{137}Cs		^{144}Ce		^{90}Sr		^{60}Co		$^{103-106}Ru$		$^{95}Zr-^{95}Nb$		TRE[a] ^{90}Y (as ^{90}Y)	
		1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963
TRM	570.8	1.4	1.8	3.9	7.2	0.45	0.61	*	0.45	2.1	7.5	1.2	6.3	3.0	4.1
	562.7	23	18	4.3	12	0.45	0.77	3.0	2.7	18	21	2.1	9.8	5.0	9.2
	552.7	29	26	5.1	14	0.77	0.65	3.8	3.4	18	26	2.2	12	6.0	13
	543.8	33	14	6.4	11	0.32	0.52	5.3	2.3	27	14	2.8	11	6.7	11
	532.0	29	13	5.9	10	0.45	0.56	4.2	2.3	24	13	2.9	9.5	4.6	8.8
	491.9	13	10	3.6	10	0.36	0.52	2.0	1.8	14	10	2.3	8.1	3.5	9.8
	475.1	9.9	9.9	2.7	11	0.36	0.74	1.7	1.8	9.9	9.9	1.8	9.0	1.7	8.9
	434.1	9.5	8.1	4.2	20	0.32	0.23	1.6	0.90	15	8.1	2.4	22	3.1	19
	381.2	5.3	5.9	1.8	9.1	0.18	0.61	0.90	0.90	6.4	5.9	0.60	9.0	0.80	8.4
	354.4	4.9	5.0	2.7	13	0.27	1.1	0.95	0.90	11	5.0	1.4	11	2.6	12
Average		16	10	4.1	12	0.39	0.63	2.3	1.7	14	12	2.0	11	3.7	10
Fort Loudoun Background Data															
TRM	604.4	1.8	2.2	4.8	10	*	0.61	*	*	5.1	9.4	3.0	6.8	2.7	5.9
	615.8	--	2.1	--	9.5	--	0.54	--	*	--	8.5	--	5.5	--	9.5
Average		1.8	2.2	4.8	9.8		0.58			5.1	9.0	3.0	6.2	2.7	7.2

[a]TRE - total rare earths minus cerium

*None detected

-- No samples taken in 1962

ORNL-DWG 64-2863A

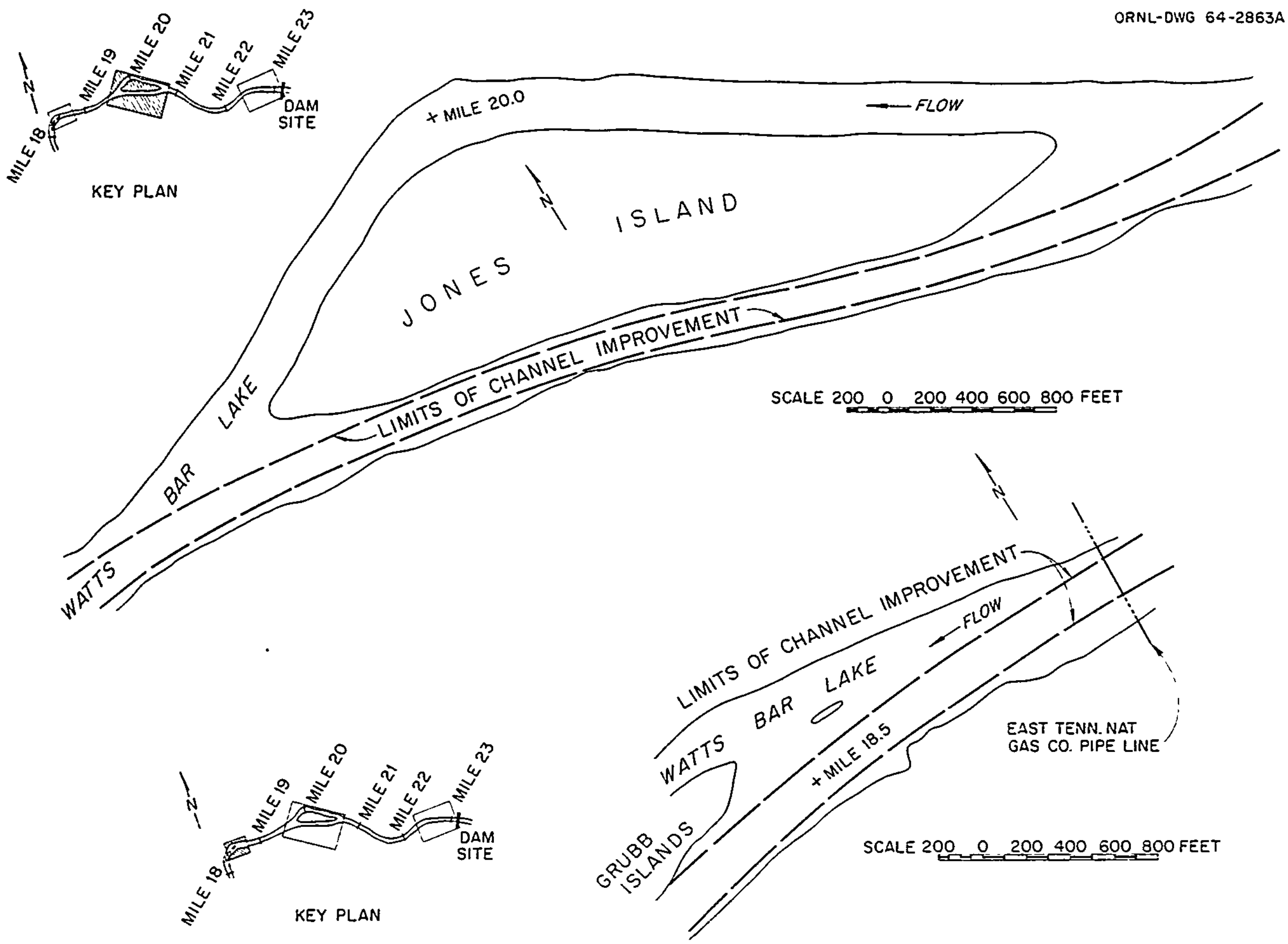

Fig. 17. Location Map–Areas of Channel Dredging near Jones and Grubb Islands

Potential Exposures from Radioactivity in the Sediments

Several special surveys have been made during the past several years and the results of the analyses of samples collected during these surveys are shown in Table 13.

Table 13. Radioactivity in Clinch River Sediment

Isotope	Emitter	Units of 10^{-6} $\mu c/gm$				
		1960 ORNL-3202[25]		1960 USPHS[26]		May 1962 ORNL[27]
		Mile 19.5	Mile 20.8	Mile 19.0	Mile 20.0	Mile 20.0-20.3
^{106}Ru	β	27.9	39.0	68.5	42.4	17.5
^{137}Cs	γ	71.2	1060	150.2	70.2	70.9
^{95}Zr-Nb				5.4	0.4	4.6
^{60}Co	γ	9.23	83.2	12.0	8.2	5.4
TRE	β	22.8	204.0			20.0
$^{89-90}Sr$	β	1.08	16.8	1.84	0.6	1.87

While there are considerable differences between specific analyses for these several samples, it appears that the concentrations of the 1960 samples by ORNL[25] at CRM 19.5 and the 1960 samples by USPHS[26] at CRM 20.0 are comparable to the 1962 samples by ORNL[27] between CRM 20.0 and CRM 20.3.

The general plan of channel improvement was to remove the bottom material from under water by a hydraulic dredge or a dipper dredge and depcsit it on nearby land areas in spoil banks or beds. The potential exposures that were visualized, depending upon the radionuclides present and their concentration and also upon the subsequent use of the spoil areas, included the following:

(1) External radiation exposure of animals or humans

(2) Radionuclides taken up by vegetation growing in the spoil areas which might find a channel of transfer to man. Media of transfer might include beef from cattle grazing upon spoil-area vegetation, tobacco grown in the spoil areas, milk from cattle grazing in the contaminated areas, or vegetables grown in the contaminated soil. Data were collected and these potential exposures were analyzed as stated later in the conclusions.

Channel Improvement Dredging

The Grubb Islands were selected as disposal areas for the material dredged between CRM 18.3 and 18.8 and Jones Island for material dredged between CRM 19.5 and 20.8. The dredging operation (hydraulic only) was begun in the Grubb Islands area in October 1962. In the Jones Island area hydraulic dredging was begun in May and completed during June, 1963. A dipper dredge was used for rock excavation immediately below the dam and for other channel work that could not be handled by the hydraulic dredge.

Finer material was excavated in the Grubb Islands reach than in the Jones Island reach and no blasting for rock excavation was required. Virtually the entire area of the Grubb Islands was used for spoil deposits and there was some extension of the island. Figure 18 shows typical spoil beds on Grubb Islands several months after they were deposited. Vegetation had begun to be established and although much of the spoil material was gravel it is expected that in a few years the area will be completely covered with vegetation. Beginning in May 1963 the hydraulic dredge worked in the

Fig. 18. Main Spoil Area - Grubb Islands. Photograph courtesy of TVA.

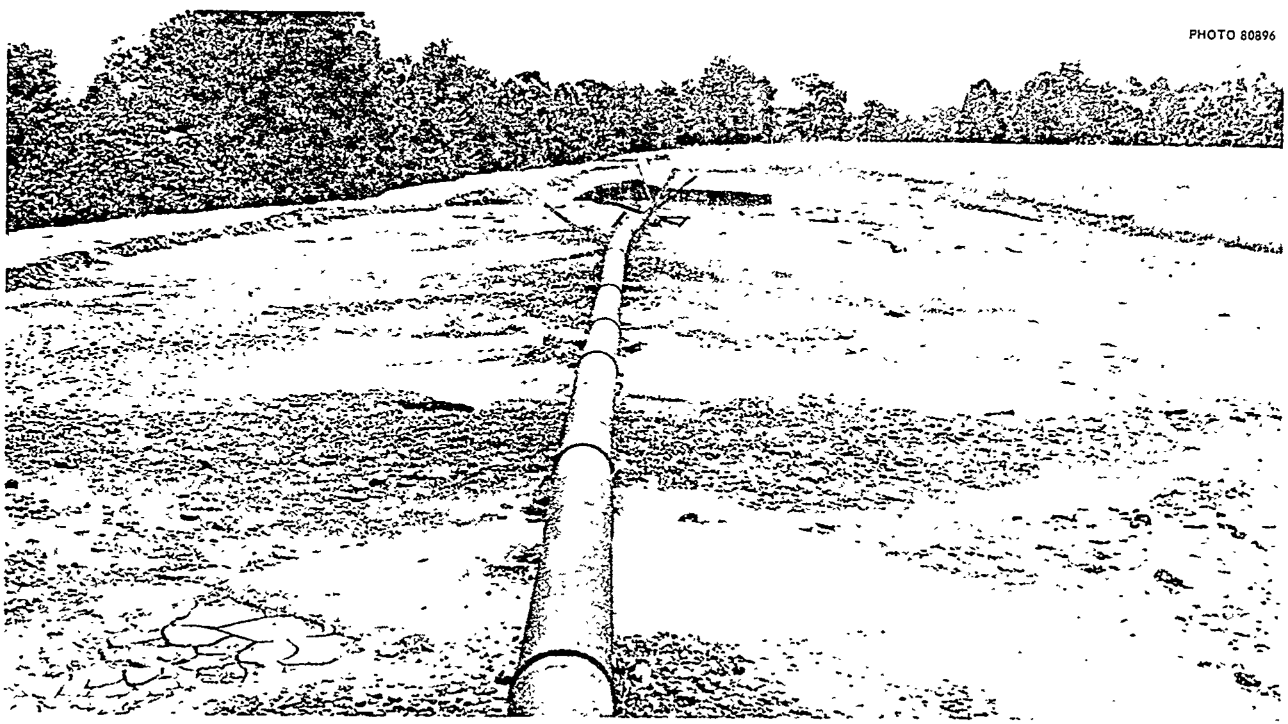

Fig. 19. North-Shore Spoil Area - Middle of Jones Island. Photograph courtesy of TVA.

Jones Island reach depositing spoil at the head of the island first. The operation then moved progressively down the channel to deposit spoil in ponds excavated on the north side of the island about the middle of the reach and at the lower end of the island. The north-shore spoil area near the middle of Jones Island is shown in Fig. 19.

Radiological Measurements and Hazards Analysis

From the beginning of dredging in October 1962 to the end of the operation of June 1963 direct radiation measurements, usually immediately above the dredged material, were made at selected points. Also radiochemical analyses of samples from the dredged materials were made at intervals, including determinations of gross beta, TRE beta, Sr beta, $^{103-106}Ru$, ^{137}Cs, ^{95}Zr-Nb, and ^{60}Co. The results of these measurements were given in the report to the Steering Committee.[6]

The water overflowing from the spoil pond back into the river carried with it much turbidity. Tailings built up in the river below the overflows and had to be subsequently removed. The spoil ponds on Jones Island retained very little fine silt or the silt that was retained was subsequently covered by sand and gravel (see Fig. 19). The sorptive capacity of sand and gravel for fission products is low as compared to that of silt. The large volumes of water used in the hydraulic operations not only carried back into the river most of the "fines" and their sorbed load of radionuclides, but also washed off "fallout" contaminants from the ground and surface vegetation.

Almost without exception the direct radiation measurements made above the spoil material were lower than the measurements made the same day over the river bank or pasture areas which were not affected by the spoil deposits. The atmospheric fallout, presumably from nuclear weapons testing, produced a background radiation greater than did the spoil deposits.

The Federal Radiation Council[28] has estimated that the annual radiation dosage from natural sources is 80-170 millirem. It recommends Radiation Protection Guides for normal peacetime operations, which would limit the annual dose to an individual in the general population to 500 mrem above background. The highest measurement made over the spoil deposits (0.052 mr/hr) would produce a total dose of only 455 mrem per year, including background. For comparison, at least three measurements made over undisturbed areas of the island indicated a dose from fallout that was this great or greater.

Of the fission products found, ^{90}Sr, ^{137}Cs, and ^{131}I are of most concern as a potential hazard to man. Because of its short half-life (about 8 days), ^{131}I could not pass from the soil to vegetation to beef animals to man in a short enough time to cause an appreciable hazard.[29] Cesium is so strongly bound to the clay that plant uptake of ^{137}Cs from soil is slight. Since ^{90}Sr is important as a potential human hazard, has a long half-life (about 25 years), and may be transferred from soil to plants to man (or animals), this radionuclide required careful review.

For purposes of the hazards analysis, assumptions were made as shown below. They are based on information from various sources to which the available references are given.

Soil depth	6-2/3 inches
Soil density	1.32 grams/cc
Soil to crop transfer	1 per cent[30,31]
Crop production	1 kg/sq meter[30,31]

Intake of grazing cow	10.9 kg/day[32]
Observed ratio (body-diet)*	0.25[33]
Calcium intake of cattle	50 grams/day/head[34]
Value for diet (calculated)	0.00049 μc ^{90}Sr/g Ca
Calcium content of meat	0.116 gm/kg[35]
Meat consumed by man	0.114 kg/day[36]

*Observed ratio is in this case, the hypothetical ability of cattle to assimilate metabolically the ^{90}Sr from its diet.

It was calculated that in the bodies of cattle the ratio of ^{90}Sr to calcium is 0.00012. For each picocurie (10^{-12} curies) of ^{90}Sr present per gram of soil, there would be transferred to the cow 14 picocuries of ^{90}Sr per kilogram of meat, and, even if it is assumed that all meat eaten is from cattle grazed on Jones Island, the resultant intake to man via meat would be less than 1.6 picocuries per day. The highest concentration of $^{89-90}Sr$ as reported by the analytical laboratory for any spoil deposit sample was 0.58 picocurie per gram. These samples were selected from the finest deposits on the surface of the spoil beds and were more concentrated than the average spoil consisting of sand and gravel. Furthermore, the soil depth of 6-2/3 inches assumed for purposes of calculation is many times the actual depth of the fine deposits on Jones Island. This maximum calculated intake is in the lowest 10 per cent of Range I designated by the Federal Radiation Council as being such that only periodic surveillance is required.

Conclusions

The excavation of material from the Clinch River channel and its deposition in spoil pools or banks on Grubb and Jones Islands resulted in the development of potentially radioactive areas. Fine solids were flushed back into the river or subsequently covered by larger material so that the exposed areas consisted almost entirely of sand and gravel, particularly on Jones Island. Vegetation has taken root and become established on the spoil deposits.

A consideration of the potential hazards led to the following conclusions:

(1) Hypothetical calculations of metabolic transport of ^{90}Sr from the spoil deposits to man <u>via</u> vegetation and beef cattle did not indicate a radiological hazard.

(2) There is no current reason to limit the use of Jones Island for beef cattle grazing. Observations should be made periodically to determine the concentration of radionuclides in vegetation which develops or is planted on the spoil beds.

(3) Any change from beef cattle grazing to dairy cattle grazing, or any proposal to grow vegetable crops for human consumption, would require further study before such use of the spoil beds should be permitted.

(4) The tobacco crop in its present location does not create a problem of strontium uptake from the spoil bed. No study was made of the possible intake from inhalation or ingestion of materials from the tobacco but it is inconceivable that a person could have as large an intake by this mode of transfer as by the consumption of beef from the area.

BIOLOGICAL PHASES

Biological investigations in the Clinch River Study are concerned primarily with immediate or long-term potential hazards to human populations. Their chief objectives are to provide data on the quantities of radionuclides in the food-chains links that lead directly to man, mainly in fish which are the primary sources of human food from the Clinch River; and to present these data in a form that can be utilized by the Subcommittee on Safety Evaluation in its analyses of Clinch River conditions.[37, 38]

In order to achieve this, joint teams from the Oak Ridge National Laboratory (ORNL) and the U. S. Public Health Service (USPHS) have cooperated in the collection and analysis of fish from the river system, more or less continuously since the study was begun in 1960. These two agencies joined in making a large collection of fish in the Clinch River during the period May 16-24, 1962. The aim was to collect 40 individuals of at least 11 common species of <u>food and game fish</u>, of which half were to be analyzed by the USPHS in Cincinnati and half at the ORNL analytical laboratories.[38]

The following year, during the period May 20-22, 1963, an additional collection of <u>food fish</u> was made from both the Clinch and Tennessee Rivers, and the work of analysis was again shared by ORNL and the USPHS. The major objectives of this additional collection were:

1. To recheck the value of the water dilution factor previously observed in comparing the radioactivity of fish from the Clinch River and from Watts Bar Reservoir.

2. To determine the relationship of methods of sample preparation (particularly the elimination of bone) to the concentrations of radionuclides found in the analyses of fish specimens.

The analytical data from the large collection of fish in 1962, from the additional collection in 1963, and from the earlier collection and analysis of river fish (1960-1962) have been tabulated and submitted to the Subcommittee on Safety Evaluation for use in the further definition of safety factors and completion of the safety evaluation by this subcommittee. A summary of the evaluation is given in a later section of this status report (see page 130 *et seq*.).

To assure that the results of analyses of fish samples by the USPHS and ORNL would be comparable, identical aliquots of more than 20 samples were analyzed by both agencies. When the means of the concentrations of ^{137}Cs and ^{90}Sr were compared and the equality of the analyses tested statistically, the results from the two laboratories were found to be equal within the 5% level of significance.

In summary, fish samples from the Clinch and Tennessee Rivers were collected and analyzed by ORNL and USPHS during 1960-1963, and the Subcommittee on Aquatic Biology collected additional data on the harvest and use of fish downstream from ORNL from both commercial fishing and sport fishing. Also, special studies which were made in the ORNL Ecology Section on (1) the use of specific activities in predicting the uptake of ^{90}Sr by white crappie, and (2) the movement of smallmouth buffalo in the vicinity of White Oak Creek are of particular interest in the Clinch River Study. The above studies are summarized or references to more detailed reports are given in the subsections below.

Fish Studies Reported by Subcommittee on Aquatic Biology

Collection and Analysis of Fish from Clinch and Tennessee Rivers

For convenience the sampling and analysis of river fish will be considered in three periods: (1) from the beginning of the Clinch River Study, January 1960, through April 1962; (2) in May 1962 and later during that year; and (3) in May 1963. These three periods correspond with the major work projects in the study of Clinch and Tennessee River fish.

During the period February 1960 through April 1962 fish were collected during various seasons and were processed and analyzed according to the current practices of the USPHS and ORNL laboratories which cooperated in these studies. The results of the analyses by the USPHS were given in preliminary reports by Friend, et al,[26,39,40] and in Status Report No. 3 on the Clinch River Study, ORNL-3370, pp. 81-85.[3] More intensive studies of fish populations and movement and accumulation of radionuclides in fish were made by the staff of ORNL during this period, and the results were reported to the Steering Committee and to the Subcommittee on Safety Evaluation.[41]

The major cooperative effort in collection and analysis of fish from Clinch River in May and later in 1962 by the USPHS and ORNL, will be described more specifically. Of a total of 390 fish caught from the Clinch River and prepared for analysis during May 16-24, and to some extent subsequently, the number of individuals of the various species of fish collected and allocations for analysis by the USPHS or ORNL are shown in Table 14.[38] Fishing was limited to one portion of the Clinch River (CRM 19.1 -

Table 14. Fish Collected and Analyzed for Use in Safety Evaluation

SPECIES	Number and Allocation of Fish Analyzed		
	USPHS	ORNL	TOTAL
White Crappie (Promoxis annularis)	20	20	40
Bluegill (Lepomis macrochirus)	20	20	40
White Bass (Roccus chrysops)	20	20	40
Largemouth Bass (Micropterus salmoides)	13	0	13
Sauger (Stizostedion v. vitreum)	13	4	17
Drum (Aplodinotus grunniens)	20	20	40
Catfish (Iactalurus punctatus)	20	20	40
Carp (Cyprinus carpio)	20	20	40
Smallmouth Buffalo (Ictiobus bubalus)	20	20	40
Carpsucker (Carpiodes carpio)	20	20	40
Golden Redhorse (Moxostoma erythrurum)	20	20	40

CRM 4.5). Although a variety of collection devices, including gill nets, hoop nets, trammel nets, and an electric shocker were used, some species were not taken in sufficient numbers to fulfill the goal of 40 individuals of 11 common species of food and game fish; the goal was not attained in the cases of largemouth bass and sauger. A majority of the large game fish were taken for analysis by the USPHS in expectation that more specimens would be forthcoming which would be handled at ORNL.

Sample Processing. A uniform sample processing scheme was developed by the USPHS and ORNL groups (see Fig. 20). The major factor governing the method used in processing a fish species was the way in which the species is normally prepared for human consumption. On this basis the flesh and bone of the bottom feeders were ground together which accounts for the significantly higher ^{90}Sr burdens

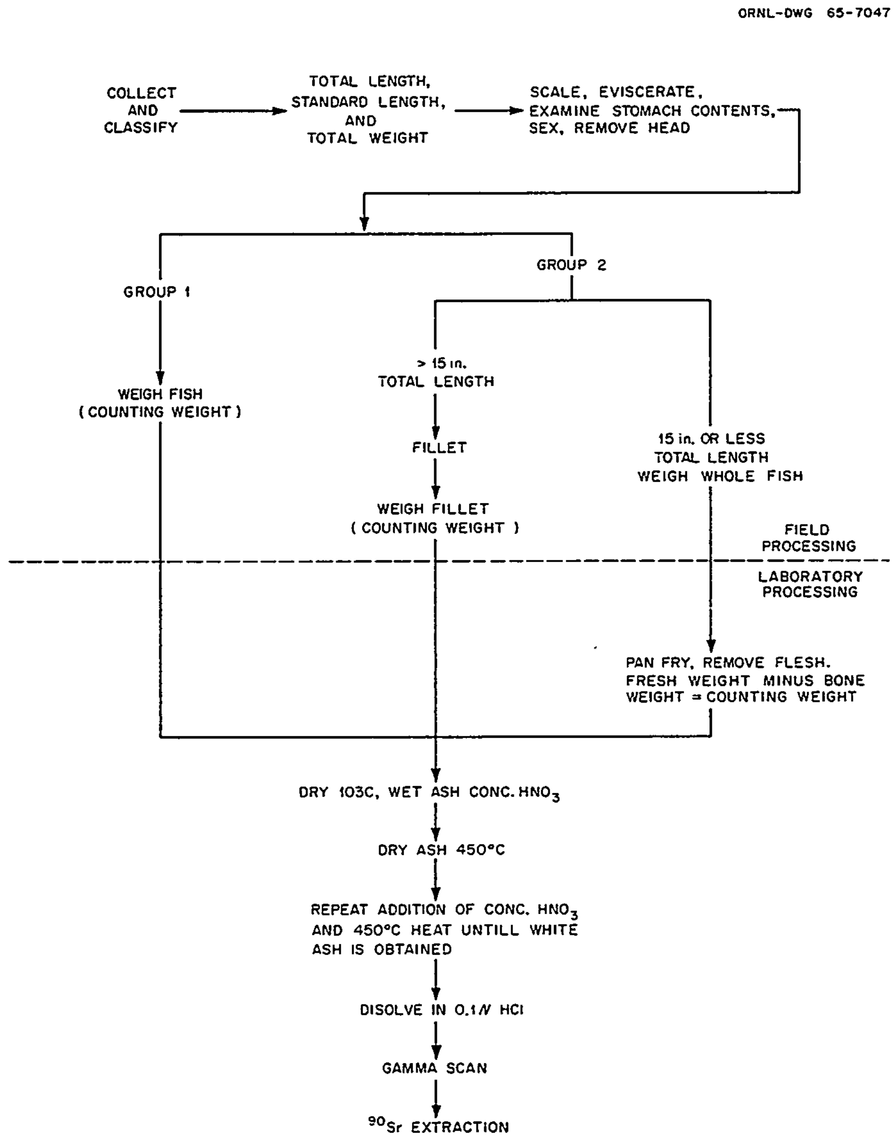

Fig. 20. Flow Sheet for Preparation of Fish Samples for Radioanalysis.

in the "Total" samples of these species (see Table 15). Thus, the exact processing method employed in the field and laboratory depended upon information regarding the fish species and method of preparation for eating and, with game fish, the size of the individual. All the fish were weighed, measured, scaled, and cleaned (including removal of the head) in the field.

The fish were divided into two groups. Group 1 included carp, carpsucker, smallmouth buffalo, and golden redhorse. Group 2 included white bass, white crappie, largemouth bass, bluegill, sauger, drum, and catfish. The catfish were channel catfish with the exception of one flathead catfish.

If fish in Group 1 were greater than 15 inches in total length they were filleted and the fresh weight of the fillet was used as the sample weight. Group-1 fish 15 inches or less in length were pan-fried and the cooked flesh was removed for analysis. The sample weight was the fresh weight minus the bone weight. Group-2 fish 15 inches or less in length were processed in their entirety, including bones. The sample weight was the fresh weight of the whole fish.

The processing of the fillets and fried flesh of Group-1 and Group-2 fish was essentially the same. All samples were dried at 103°C, wet ashed with concentrated HNO_3, and ashed with heat at 450°C. The ash was further treated with concentrated HNO_3 and heated until a white ash was obtained. This was dissolved in 0.1 N HCL with a final volume of 25 to 1000 ml, depending upon the size of the fish. Samples were scanned for gamma emitting radionuclides and strontium was precipitated for ^{90}Sr determinations.

Table 15. Concentration of Radionuclides in Clinch River Fish

(pc/kg fresh weight)

Fish Species	SAMPLE PERIOD	^{90}Sr Flesh	^{90}Sr Total[a]	^{137}Cs Flesh	^{137}Cs Total[a]	^{106}Ru Flesh	^{106}Ru Total[a]	^{60}Co Flesh	^{60}Co Total[a]
Carp	1960 - 1962	(17)[b] 500 ± 140	(40) 5100 ± 1700	(71) 510 ± 57	(39) 560 ± 79	(69) 170 ± 18	(39) 290 ± 78	(67) 66 ± 6.1	(39) 49 ± 9.9
	1963	(20) 91 ± 22		(20) 320 ± 110					
Carpsucker	1960 - 1962	(18) 540 ± 190	(39) 940 ± 120 (39) 4800[d]	(122) 1200 ± 460	(37) 640 ± 67	(22) 120 ± 30	(37) 56 ± 16	(22) 120 ± 19	(37) 32 ± 6.8
	1963	(20) 22 ± 4.4		(20) 460 ± 82					
Buffalo	1960 - 1962	(3) 240 ± 89	(30) 830 ± 110	(5) 480 ± 94	(30) 590 ± 92	(5) 110 ± 32	(30) 150 ± 38	(5) 78 ± 21	(30) 32 ± 6.8
	1963	(20) 43 ± 14		(21) 560 ± 84					
Sight Feeders[c]	1960 - 1962	(109) 180 ± 83		(126) 680 ± 120		(127) 120 ± 32		(127) 22 ± 11	

[a]Total fish consists of flesh and bone.

[b]Parenthetical values are numbers of fish analyzed.

[c]Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

[d]Includes four carpsuckers (composited) collected at CRM 19.6.

Table 16. Concentration of Radionuclides in Flesh of Tennessee River Fish

(μμc/kg fresh weight)

Fish Species	Sample Period	^{90}Sr	^{137}Cs	^{106}Ru	^{60}Co
Carp	1960-1962	(13)[a] 120 ± 33	(14) 180 ± 55	(14) 80 ± 27	(14) 71 ± 17
	1963	(20) 5.1 ± .75	(19) 61 ± 17		
Carpsucker	1960-1962	(10) 99 ± 28	(10) 130 ± 27	(10) 69 ± 23	(10) 62 ± 18
Buffalo	1963	(20) 8.9 ± 2.9	(20) 73 ± 12		
Sight Feeders[b]	1960-1962	(24) 250	(24) 170	(24) 48	(24) 66

[a]Parenthetical values are numbers of fish analyzed.
[b]Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

The same processing procedures were used for the fish collected in May 1963 except that, for comparison with the 1962 analyses, the bone was not included in the processing of selected portions of the fish. The purpose was to determine what influence, if any, the inclusion of bone in the preparation of fish would have on the concentration of radionuclides.

Analytical Results. Results of analyses of fish collected from Clinch River in 1960-1962 are shown in Table 15 [42] and the fish collected from the Tennessee River in 1960-1962 are shown in Table 16.[42] These tables also give the results of analyses of carp, carpsucker, and buffalo collected from the Clinch River and of carp and buffalo obtained from the Tennessee River in May 1963. In the calculations for safety evaluation studies, estimates were made of the intake of radionuclides by a man who consumed, at a specific assumed rate, Clinch River fish or Tennessee River fish containing the concentrations of radionuclides shown in Tables 15 and 16 respectively.

Data on Harvest and Use of Fish

Because of the observation mentioned earlier that inclusion of bones in the preparation of bottom feeders for eating resulted in significantly higher ^{90}Sr burdens in these species, the Subcommittee attempted to develop further information on the total quantities of bottom feeding fish harvested from Watts Bar Reservoir. These data were compared with the total East Tennessee harvest, including the relative amounts shipped to various market centers, and were summarized in Progress Report No. 2 of the Subcommittee on Aquatic Biology.[38] Data on the commercial fish harvest are shown in Table 17 and the distribution of commercial fish from East Tennessee is shown in Table 18. Of particular interest is the fact that all of the carp were marketed within the state of Tennessee, in the cities of Nashville and Memphis. *

The Subcommittee was not able to obtain as definite information about sport fishing and the data on capture and use of game fish are meager in comparison with those on commercial fishing. The progress report [38] summarizes and tabulates what data were obtained regarding sport fishing and game fish. Although incomplete these data give some indication of the distances from which fishermen come to fish in the Tennessee River, the total number of various species of fish caught by fishermen who had traveled various distances to fish in Watts Bar Reservoir and the Clinch River, and the area of maximum consumption of Clinch River fish caught either in the Clinch River or in Watts Bar Reservoir. For example, the Tennessee Game and Fish Commission conducted a partial creel census on Watts Bar Reservoir from March 1 through May 15, 1961 and during this period cards were issued to 1213 fishing parties. The returns, for only a part of the year, represented approximately 2444 fishermen, of which 3.85% were not residents of Tennessee. Using the data derived from ORNL fish tagging studies performed in 1962 an estimate of the area of maximum consumption of Clinch River fish was attempted. It appeared that, of the fish tagged, the greatest percentage (72.5%) were utilized within 50 miles of the site of capture.

One small enquiry about use of fish by people in this area was made by means of a questionnaire filled out at a country-wide meeting of home demonstration clubs. Of 53 individuals who answered the questions: 39 stated that they did use fish caught from local lakes and rivers while 14 said they did not; the kinds of fish used included bass, bream, crappie, carp, catfish, and others but the majority (30) listed crappie;

Table 17. Commercial Fish Harvest from Watts Bar Reservoir and the Total Harvest from East Tennessee

	Carpsucker	Carp	Smallmouth Buffalo
Watts Bar Reservoir	15,608 lbs	23,667 lbs	161,304 lbs
East Tennessee	61,676	135,016	327,302
Fish Dilution Factor*	3.95	5.70	2.04

$$*\text{Fish dilution factor} = \frac{\text{Lbs of East Tennessee Fish}}{\text{Lbs of Watts Bar Fish}}$$

Since there is no commercial fishery on the Clinch River, an average water dilution factor of 5.6 may be applied to the Watts Bar fish in addition to the fish dilution factor.

Fish and Water Dilution Factor	22.12	31.92	11.37

Table 18. Distribution of Commercial Fish from East Tennessee to Consumer Markets*

Species	Consumer Markets	Percentage
Smallmouth Buffalo	Chicago, Illinois	40
	Louisville, Kentucky	30
	Memphis, Tennessee	30
Carp	Nashville, Tennessee	50
	Memphis, Tennessee	50
Carpsucker	Chicago, Illinois	10
	Nashville, Tennessee	45
	Memphis, Tennessee	45

*Estimates from Mr. Harold LaTendresse of Johnny's Fish Company, Knoxville, Tennessee as to methods of preparation a majority indicated pan-fry and fry in deep fat, but broil, bake, and other methods were also listed; and estimated quantities of local fish used per family averaged 10 pounds per month. Regarding frequency of use, 19 said that they served fish caught from local waters about once per week, and 20 said once per month.

Use of Specific Activities in Predicting the Uptake of ^{90}Sr by White Crappie (*Pomoxis annularis*)

In Status Report No. 4 the section on biological studies reported a series of determinations of stable strontium (Sr) in white crappie (*Pomoxis annularis*) flesh and bone.[43] These determinations were completed and the observed activities were compared with calculations of expected body burdens of ^{90}Sr in these fish. Samples were taken at monthly intervals to determine whether there were seasonal changes in Sr concentrations in fish flesh and bone. Also, bone and flesh samples were analyzed for Ca, and bone samples were analyzed for ^{90}Sr.

The Ca and Sr concentrations in fish flesh and bone were practically constant throughout the year as shown by the average annual concentrations and standard deviations given in Table 19. Relatively constant concentrations were expected in fish bone because of the long biological half-life of Ca and Sr in bony tissue. No seasonal variation was detected in the Ca and Sr concentrations in the fish. This is probably a reflection of the relative constancy of these elements in Clinch River water as reported earlier from stable-chemical analyses of water samples.[3]

Of particular interest was the fact that the average concentration of Sr in fish flesh was essentially the same as that of river water. This concentration factor of 0.01

Table 19. Calcium and Strontium Concentrations in White Crappie Flesh and Bone and in Clinch River Water

White Crappie	No. of Samples	Ca	Concentration Factor	Sr	Concentration Factor
Flesh (μg/g)	113	135 ± 2.43	6.34	0.068 ± 0.035	1.01
Bone (mg/g)	105	365 ± 7.77	17,136	0.268 ± 0.064	3941
Clinch River Water (μg/g)		21.3 ± 3.0		0.068 ± 0.0094	

All values are average annual concentrations ± one standard deviation

$$\text{Concentration Factor} = \frac{\text{concentration/g tissue}}{\text{concentration/ml water}}$$

Table 20. Specific Activity Relationships of ^{90}Sr between Clinch River Water and White Crappie Bone (Calculation)

$$\frac{^{90}\text{Sr}}{\text{Sr}} \text{ Water} = \frac{^{90}\text{Sr}}{\text{Sr}} \text{ fish bone}$$

$$\frac{4.3 \times 10^{-3}\ \mu\mu\text{c/ml}}{6.8 \times 10^{-2}\ \mu\text{g/ml}} \cong \frac{1.446 \times 10^{1}\ \mu\mu\text{c/g}}{2.684 \times 10^{2}\ \mu\text{g/g}}$$

$$6.23 \times 10^{-2}\ \mu\mu\text{c}/\mu\text{g} \cong 5.39 \times 10^{-2}\ \mu\mu\text{c}/\mu\text{g}$$

was far less than that of 20,000 to 30,000 times, which was reported from an earlier study of fish in White Oak Lake.[44] In fact, it is difficult to understand fully such a large concentration factor since (as shown in Table 19) Sr was concentrated only by a factor of 3941 times in bone, the tissue that would be expected to have the highest concentration.

To test whether specific activity ratios of ^{90}Sr in fish tissue were related to those in water, the concentrations of ^{90}Sr and Sr in bone were compared by use of the proportion given in Table 20. The data on ^{90}Sr concentrations in water, obtained from the Applied Health Physics Section,[45] were for the period of time these fish lived in the river water; and data on concentrations of stable Ca and Sr in Clinch River water are shown in Table 19.[3] There was relatively good agreement between the specific activities in fish bone and in water. These comparisons suggested that for any constant release of ^{90}Sr to the river, the consequent ^{90}Sr content of fish bone could be predicted.

The studies of the applicability of specific activities to predict ^{90}Sr concentrations in fish tissue were supplemented with investigations of the biological half life of Sr in white crappie. Knowledge of the biological half life in fish flesh or bone will permit calculations of the rate at which ^{90}Sr concentrations in fish tissues will respond to changing environmental concentrations of ^{90}Sr. Using ^{85}Sr and whole-body counting techniques no significant loss of Sr from bone was detected. On the other hand the biological half-life of Sr in the flesh of different individual fish has ranged from 12 to 48 minutes, much shorter than was anticipated.

Measurement of the turnover of ^{85}Sr in fish flesh was complicated by the rapid deposition and high concentration of Sr in fish bone, which literally masked the Sr in soft tissues. This difficulty was circumvented by constructing a fish-holding assembly, which was inserted into the detector chamber of a small-animal whole-body counter. Water was pumped through the assembly during experiments to remove excreted ^{85}Sr and to provide a supply of oxygenated water.

The long biological half-life of Sr in fish bone suggests that ^{90}Sr concentrations in the bone of river fish will reflect the average environmental concentrations during the lifetime of the fish. The short biological half-life of Sr in flesh means that fish flesh will reflect very quickly any changes in ^{90}Sr concentrations in the environment. This information is important in connection with the expected operating mode of Melton Hill turbines and releases of radioactivity from White Oak Creek.

Movement of Smallmouth Buffalo (*Ictiobus bubalus*) in the Vicinity of White Oak Creek

In the White Oak Creek embayment between White Oak Dam and the mouth of the creek, the levels of radioactive contamination in the water and bottom sediments are generally much higher than in the river. This portion of the embayment is in the controlled area which is subject to patrol, and fishing in this reach is prohibited. Fish are free, however, to move from the controlled area of White Oak Creek embayment into the Clinch River where they may be taken by fishermen and used for food.

This possibility has caused some concern that the movement of contaminated fish into the open river might contribute to the radiation exposure of the adjacent population. Some pertinent information with a direct bearing on this problem was obtained in the course of a study of the smallmouth buffalo (Ictiobus bubalus) population in the Clinch River and adjacent downstream portions of the Tennessee River.[46]

A part of the objective in this study was to test the hypothesis that fish residing in White Oak Creek would deposit radionuclides in their scales in sufficient quantities to be detected by gross beta or gamma counting and autoradiography. On the other hand fish residing in less contaminated areas would not be expected to have readily detectable radioactivity in their scales. It was further hypothesized that fish having once lived in White Oak Creek would retain radioactivity in their scales because of the long biological half-life of mineral elements in bony tissue.

Fish scales are used in conventional age and growth studies to interpret past growth histories of fish because growth of the scales is proportional to growth of the fish. If a fish spent any appreciable amount of time in White Oak Creek embayment and radioisotopes were deposited in the scales, it should be possible to use the growth proportionality between scales and fish and, from radioactive rings in the scales, determine when a fish entered the embayment and how long it stayed there.

Eleven smallmouth buffalo were found that had sufficient radioactivity in their scales to produce autoradiograms. Of these fish, eight were caught in White Oak Creek embayment, one at CRM 21.7, one at CRM 16.0, and one at TRM 542, 12 mi upstream from Watts Bar Dam. There were 146 fish in the Clinch River–White Oak

Creek embayment sample and 1271 fish from Watts Bar Reservoir between TRM 539 and TRM 544; and of these 6.8% and 0.08%, respectively, contained sufficient radioactivity for autoradiography.

In the ORNL report on this study,[46] the case history, including the movement, of each of the 11 fish examined autoradiographically is discussed separately. Of the 11 fish, there were 16 instances where movement occurred between noncontaminated and contaminated areas. Twelve of these moves coincided with resumption of growth at the time of annulus (scale-ring) formation which would indicate that the majority of the moves occurred during late winter or early spring. None of the fish left the area where it was spawned before it was two years old. The area of spawning is referred to as being either contaminated or noncontaminated. The discussion and an autoradiogram for one of the 11 fish (smallmouth buffalo No. 8) taken from the ORNL report are given below.[46]

"Smallmouth buffalo[8](Fig. 21) was hatched in the spring of 1957 in a noncontaminated area. It entered a contaminated area immediately after formation of its second annulus, probably in the spring of 1959. This fish was 271 mm long at formation of the second annulus. It remained in the contaminated area until some time during the winter of 1959–1960 when it left the contaminated area at a length of 304 mm. The animal was in a noncontaminated area until its capture on June 29, 1962, just prior to the formation of its fifth annulus. This fish was captured at CRM 16.0 at a length of 460 mm and weight of 1,410 g. Scales averaged 6 beta cpm per scale over background at capture." The autoradiogram of scales from this fish is shown in Fig. 21.

The data from this and the other 10 fish indicate the extent of smallmouth buffalo movements between White Oak Creek embayment, the Clinch River, and the

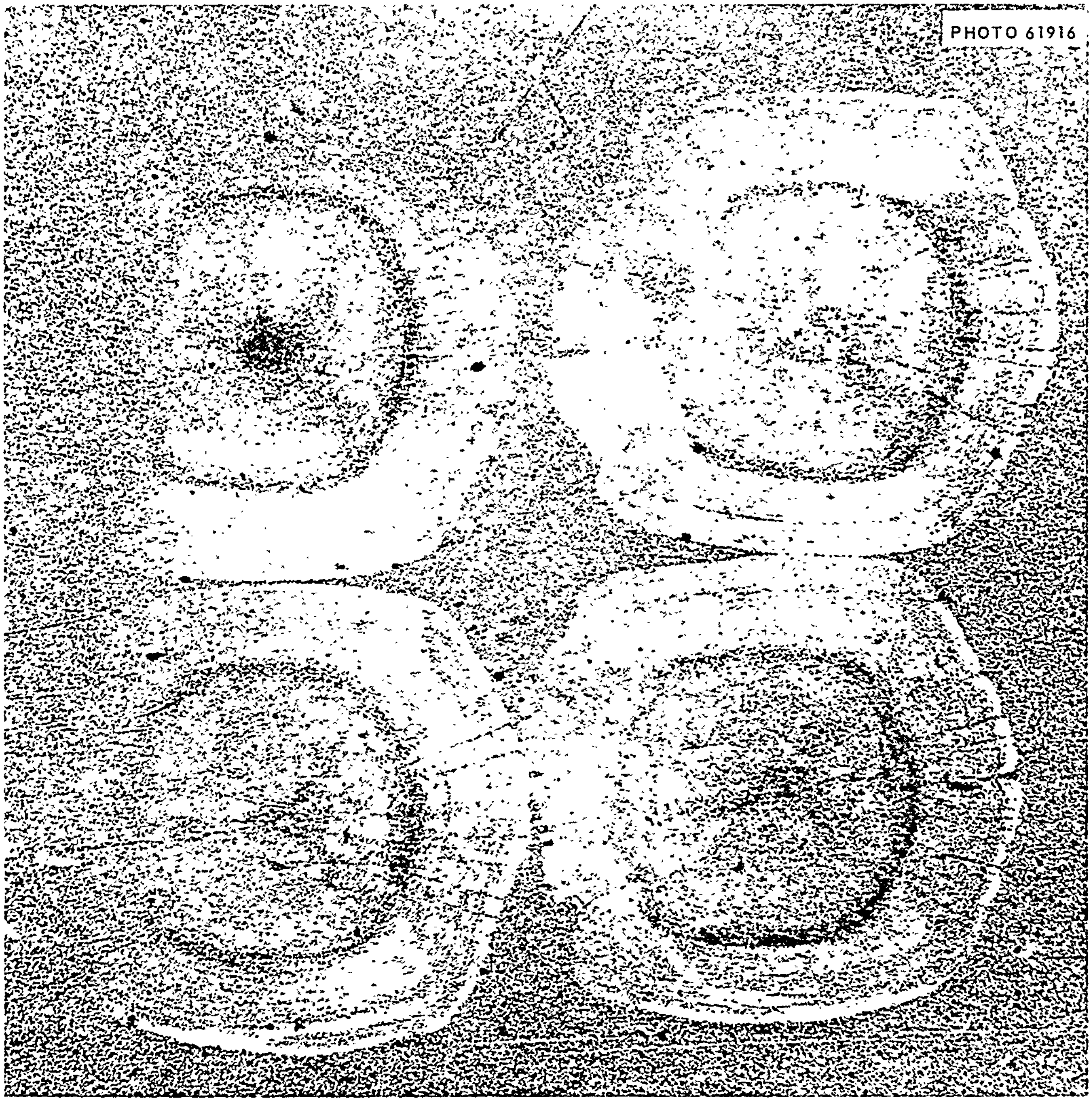

Fig. 21. Autoradiogram of Scales from Smallmouth Buffalo 8

Tennessee River portions of Watts Bar Reservoir. Use of radioactive rings permits interpretation of the history of fish movement in considerably more detail than from conventional tagging methods. Conventional tagging techniques permit the recovery of fish movement data only at points in time coincident with initial tagging and subsequent recaptures. Radioactive scale rings provide a continuum of information concerning the movement of the fish with respect to radioactive areas.

This study of the movement of this commercially important species of fish is significant because a new technique of study was developed and found to be superior to conventional tagging methods. A hazards analysis, if made, relating fish movements to the spread of radioactive contamination should be interpreted in the light of information on the biological half life of particular radionuclides in fish tissues.

HYDROLOGIC MEASUREMENTS AND ANALYSES

Cooperative Program of USGS

During the period covered by this report, April 1962 to December 1963, the U. S. Geological Survey (USGS) has continued its cooperative program of routine measurements and special observations relative to surface water hydrology in the Oak Ridge area. The USGS program in this area was summarized in Status Report No. 4 on the Clinch River Study.[47]

A primary role of the USGS in its relations with the Clinch River Study Steering Committee is to provide the basic data and the analyses that are necessary for a clear understanding of the dispersal of the radioactive wastes discharged into the Clinch River. It was for this purpose that the network of 8 gaging stations was established and 24 partial record stations were operated as described in Status Report No. 4. As explained in previous reports, the information collected at each of the stream gaging stations generally includes a continuous record of stage, discharge, and temperature. A temperature recording station at CRM 5.5 was operated for a temporary period (1961-1962) by ORNL and USGS personnel to obtain information on water temperatures at various depths in the river. In addition two special investigations conducted by the USGS jointly with other agencies engaged in the Clinch River Study are reported in subsequent subsections, namely, "Dispersion Investigations" and "Dye Tracer Studies - Summer 1963."

During the period covered by the present report there has been one major change in the gaging station network. The gaging station on Clinch River at Scarboro, Tennessee was discontinued on September 30, 1962 as a result of the filling of the reservoir above Melton Hill Dam to spillway level. This station was replaced by a station at Melton Hill Dam for which records of headwater elevations are furnished by the TVA and a continuous record of discharge through the spillway gates is provided by the USGS to all interested committee members and staff personnel of the Clinch River Study.

Dispersion Investigations

Several studies have been made or are in progress for investigation of the effects of power waves on dispersion. Most of this work has been undertaken to define the influence of power releases from Melton Hill Reservoir on the transport of radionuclides in the river system downstream.

Assuming that present methods of releasing radioactive materials from White Oak Lake are continued, a reservoir of radioactivity will accumulate in Clinch River waters in the vicinity of White Oak Creek during periods of no flow from Melton Hill Reservoir. During periods of power releases from the reservoir, this radioactive accumulation will be swept downstream in a cloud. The concentration of radioactivity in the cloud may be considerably higher than present levels.

Preliminary Observations in the Clinch River

During the summer of 1962 two periods of essentially no flow occurred in the lower Clinch River. During each of these periods water samples were collected to determine the dispersion of releases from White Oak Creek into the still pool.

This work, done by personnel of the ORNL Applied Health Physics Section, indicated that the releases from White Oak Lake tended to be dispersed upstream toward Melton Hill Dam during the period of no flow in early August. Although the exact cause of the upstream movement is not known, it could have been adverse hydraulic gradients or upstream winds which were observed at times during this period. Winds probably caused an observed upstream movement of debris on the water surface.

A period of no flow in August 1962 was established to allow the closure of a diversion channel at Melton Hill Dam. Personnel of TVA advised the investigators on the Clinch River Study when flow over the spillway would be resumed; and that the start of flow would be somewhat similar to a power release from the reservoir.

At the start of flow a fluorescent dye, Rhodamine B, was injected as a tracer, and movement of the tracer was observed for 3 days from the section of injection at the mouth of White Oak Creek (CRM 20.8) to Gallaher Bridge (CRM 14.4). The results of this test were considered as preliminary information and were used in the planning of the comprehensive series of dye-tracer studies in August 1963. The detailed results of the test were analyzed by the U. S. Geological Survey in Washington, D. C.

Laboratory Flume Study

A series of distorted-model dispersion tests were made in a flume operated by the U. S. Geological Survey at the Hydraulics Laboratory, National Bureau of Standards, Washington, D. C. These tests were undertaken to acquire immediate qualitative information on the effects of power waves on dispersion of radionuclides and to develop numerical methods of describing the diffusion process.

The flume was 150 ft long, 3.5 ft wide, and 1.5 ft deep, divided into a 1.5 ft-wide test channel and a 2.0 ft-wide bypass channel. The flume could be tilted to any desired gradient. Discharges were obtained from a 300,000-gal recirculating system and depths in the flume were controlled by a tail gate. A double-notch weir plate was installed in the forebay so that the flow could be instantaneously diverted from the bypass channel to the test channel by operation of simple slide gates at the weir.

The discharge, bed slope of the flume, water depth for no-flow conditions, and bed roughness were adjusted to conform, at distorted scales, to anticipated conditions in the Clinch River. Horizontal distortions were approximately 250:1, and vertical distortions were about 20:1.

In the test procedure, the desired flume slope and water depth for no-flow conditions in the test channel and the desired discharge in the bypass channel were established. The power wave was created by suddenly diverting the established discharge from the bypass to the test channel. At the instant the wave was created, a tracer was released. In most tests the tracer was a mixture of dissolved potassium chloride, water, and ethyl alcohol, the density of the solution being the same as of the laboratory water. The variation of salt concentration with time was measured continuously at several points in the flume, using recording conductivity bridges. At the completion of a wave test, a steady-flow test was run. The discharge, slope, tracer, and water depth were the same as for the wave test.

The variation of concentration with time and distance in the model for one complete test is shown in Figure 22. The results from this test indicate that

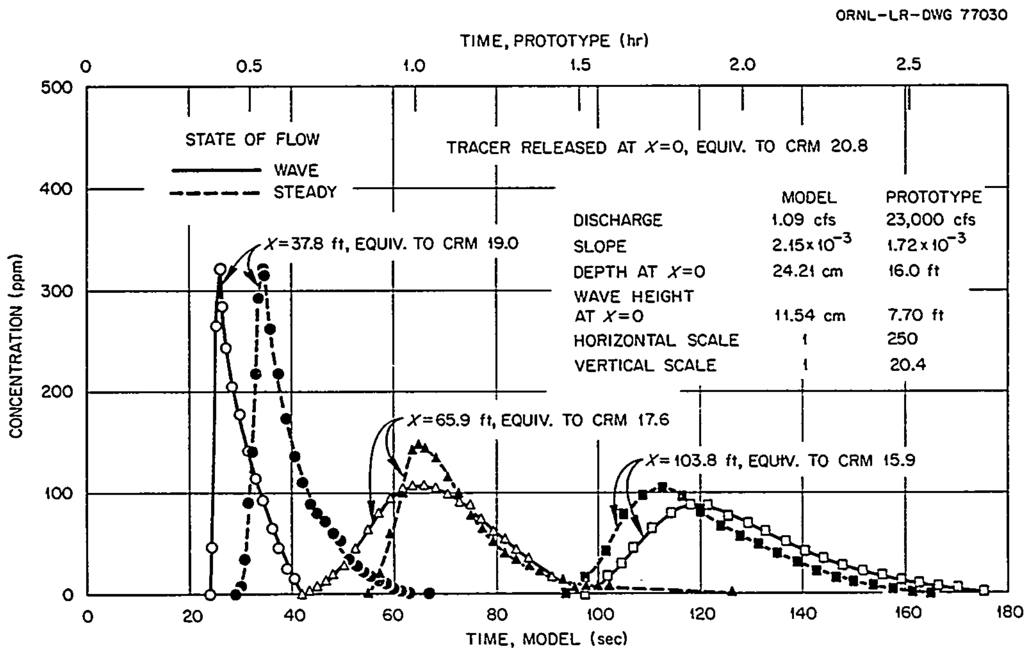

Fig. 22. Variation of Tracer Concentration with Time and Distance

the dispersion process due to power releases is not greatly different from that for steady flow, especially at sections that are considerably removed from the tracer-injection section. At equivalent CRM 15.9, about 1 1/2 miles upstream from the intake for the ORGDP water plant, the peak concentrations and the durations of the tracer cloud for the wave and steady-flow conditions were about the same.

As a check on model similitude, wave height and travel time for the model were compared to height and times predicted by TVA for the Clinch River. For flow conditions shown in Fig. 22 the equivalent wave height was 7.72 ft, as compared to the predicted height in the river of 7.70 ft. The equivalent travel time for the scale ratios indicated in Fig. 22 was about 3% greater than the time interpolated from information furnished in reports by the TVA.

Dye-Tracer Studies - Summer 1963*

Introduction

Past experience has shown that the dilution afforded by the flow of water in the Clinch River has been effective in reducing the concentration of radioactive wastes from White Oak Lake to acceptable levels. The radionuclide concentrations in river water at the Gallaher Bridge sampling station (CRM 14.4) during the past few years has been less than 1% of the maximum permissible occupational concentration in water.[48] Drastic changes in the flow regime of the Clinch River after the Melton Hill power station is put into operation are expected. During the summer months of 1963 a series of tracer studies was conducted to investigate the effects

*Based on a report by B. G. Frederick (USGS), F. L. Parker (ORNL), and P. H. Carrigan, Jr. (USGS), presented at open meeting of the Clinch River Study Steering Committee, December 4, 1963.[49]

that power releases from Melton Hill Reservoir will have on the dispersion in Clinch River of radioactive releases at White Oak Dam.

Although of relatively short duration, these studies were a rather large undertaking involving the efforts of between 25 and 50 workers either on the Clinch River or in supporting roles. It is impracticable to name the individuals who contributed, but the studies were a truly cooperative effort by the U. S. Geological Survey, Oak Ridge National Laboratory, U. S. Public Health Service, Tennessee Department of Public Health and Tennessee Valley Authority.

The studies were made by means of a nontoxic, red-colored fluorescent tracer, either Rhodamine B or Pontacyl Brilliant Pink B dye, and releases of water over the gated spillway of Melton Hill Dam at rates of discharge to simulate the expected power releases through the turbines after the electric-generating equipment is installed and operative. The estimated summer-discharge pattern of power releases at Melton Hill Dam is shown in Fig. 23.

It was assumed: (1) that proper gate releases at the dam would satisfactorily simulate power releases in the temperature and hydrologic effects produced below the dam and (2) that the tracer material (the fluorescent dye) would behave in a manner similar to radioactive wastes in the water phase of the river-study system. From preliminary surveys of temperatures at various depths in Melton Hill Reservoir upstream from the dam and also consideration of the probable conditions that might affect the dye, it was believed that the above two assumptions were acceptable.

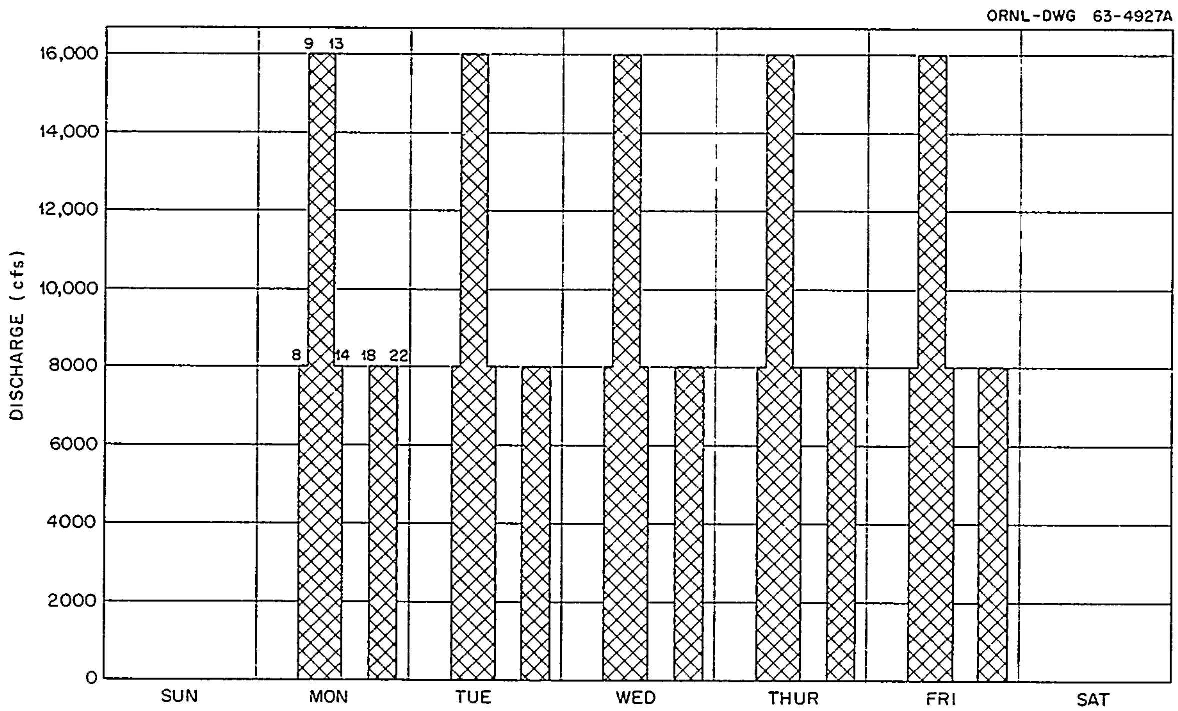

Fig. 23. Estimated Summer Discharge Pattern at Melton Hill Dam

Limited preliminary investigations also indicated that the dye would not be visible in any public water supply and would not be adsorbed on condenser tubes in the Kingston steam plant of TVA.

Following release of the fluorescent dye into the creek or river, the concentrations of the tracer in the water were determined at fixed observation sections in the stream. These determinations were made and recorded, either continuously or periodically, using Turner Model 111 Fluorometers equipped with flow-through doors and recorder attachments.*

One of the principal advantages of fluorometric instrumentation is the extremely low concentrations of fluorescent materials that may be measured. The instrument is capable of reliable detection, under field conditions, of dye concentrations ranging from 0.1 ppb (parts per billion) to about 400 ppb.

Experimental Releases and Field Observations of Dyes

Two tests were conducted under summer power-release conditions. Regulated flow in the Clinch River below Melton Hill Dam was provided, through cooperation of the Tennessee Valley Authority, by manual operation of the spillway gates at the dam. The rates of discharge at Melton Hill Dam and the weekly time schedule of intermittent power releases were as shown in Fig. 23.

In the preparatory stages of the tracer studies and prior to the power-release tests, three preliminary tests were conducted to determine the quantity of tracer needed

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*Procured from G. K. Turner Associates, 2524 Pulgas Avenue, Palo Alto, California.

in succeeding tests and to assess the reliability of the instrumentation under field conditions. Two of the preliminary tests were made with steady releases of water from Melton Hill Reservoir and a third with simulated power releases. Subsequent tests were run with the typical pattern of summer water releases (see Fig. 23)[50] and with Rhodamine B dye as the tracer.

The two tests conducted under summer power-release conditions differed largely in the duration of injection of the tracer.

1. In the first test the dye was injected continuously for 24 hr into the nappe of the effluent over the weir at White Oak Dam.

2. In the second test the dye was injected continuously for one week -- from 2:00 a.m., Wednesday, August 21, to 2:00 a.m., Wednesday, August 28, 1963 -- into the nappe at White Oak Dam.

Calculated predictions of the dispersion expected during the tests under the various conditions of water flow and dye releases were made by methods mentioned later under "Theoretical Investigations." The predicted values were compared with the actual concentrations which were observed during and following each of the first and second tracer tests. The dispersion with time which occurred in the Clinch River during the two tests was very similar to the predicted dispersion of radioactive wastes from White Oak Creek after power generation is begun at Melton Hill Dam.

In the first test (see "1" above) it was the intention that dye be injected continuously for a 24-hr period at White Oak Dam at a rate proportional to the discharge from White Oak Lake and that during this period typical releases of water from Melton Hill Reservoir be made. The dye-release apparatus did not perform properly, and the injection was not accurate, but valuable data were obtained at several points along the Clinch River. A comparison of predicted and observed dye concentrations at CRM 16.2 for four days following the release of dye in the first test has been shown graphically by a chart in the radioactive waste progress report for May-October 1963.[50]

In addition to fluorometer measurements, the progressive dispersion of the dye in the creek and river was observed visually as the intermittent releases of water were made. The spread of the red dye was easily followed by viewing from the ground or by aerial photography in the vicinity of White Oak Creek. The progressive spreading of the dye pulse in the stilled water was observed in aerial photographs taken on Wednesday afternoon, August 28. A sketch of the results is shown in Fig. 24 which indicates the extent of visible dye after the cessation of water release in the river.

At the time when flow suddenly began in the river, a rapid rise in the river water level occurred at the mouth of White Oak Creek. As a result of the increased water level, a reverse hydraulic gradient was created in the creek from its mouth to White Oak Dam; and an upstream flow occurred. For a considerable part of the time when water was being released through Melton Hill Dam, there was little or no flow from the creek

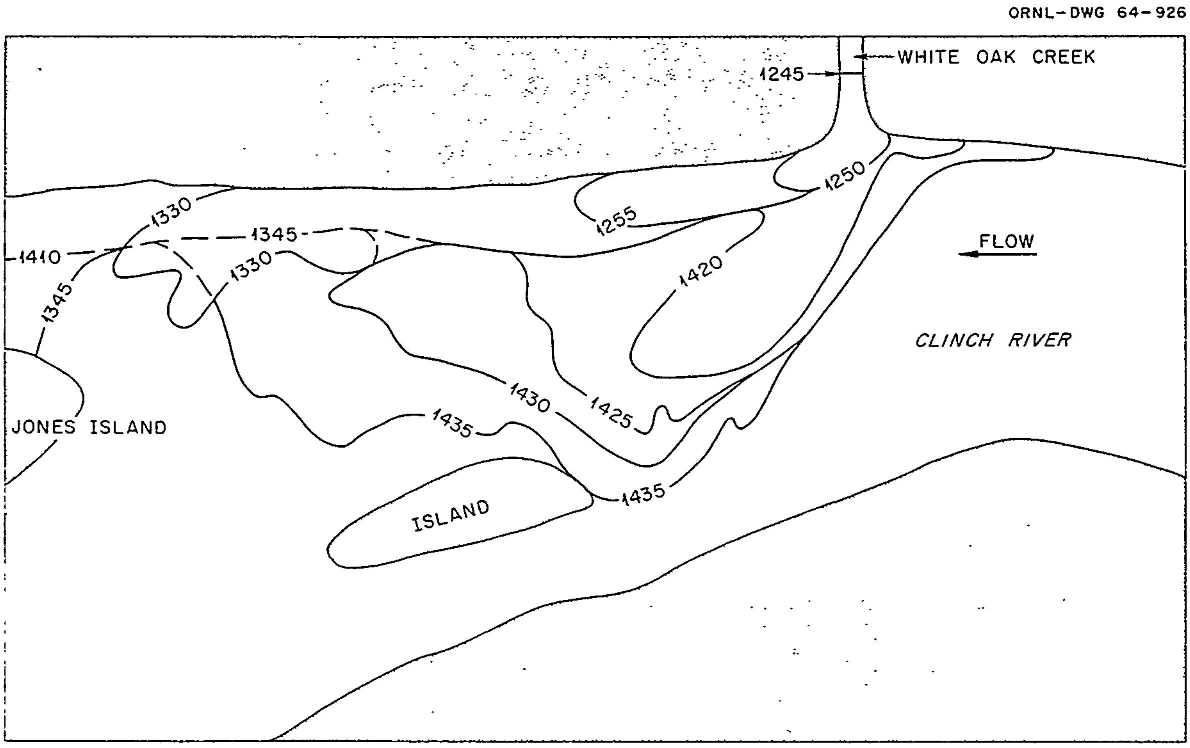

Fig. 24. Sketch of Time Development of Dye Front at and After the End of a 16,000 cfs Release in Clinch River at Mouth of White Oak Creek

into the river. When the release was stopped, the water level in the river quickly dropped, and a sudden rush of water from the White Oak Creek embayment ensued. A discrete pulse of dye accompanied this rush of water from the creek. Because there was no flow in the river, the dye mass remained close to the mouth of the creek. At the beginning of the next Melton Hill release, the slug of dye in the river was swept downstream as a reasonably intact mass, and flow up White Oak Creek again occurred. As the dye was moved downstream the color density rapidly decreased, and became practically indiscernible after about 7 miles of movement.

In the second and main test, dye was injected continuously into the effluent at White Oak Dam for 7 days, August 21-28, 1963. During this period the discharge from White Oak Lake was 6.9 cfs ± 2% (1.17×10^7 g/min*), and, except for the first 4-5 hrs at the start of the test when the injection pump did not operate properly, dye was injected at a rate of 32 g/min, resulting in an average dye concentration in White Oak Creek just below White Oak Dam of 2.73×10^3 ppb (parts per billion).

Dye Concentrations, Predicted and Observed. - Samples for dye analyses were taken every two hours from White Oak Creek below White Oak Dam during the third test, and continuous observations of the dye concentrations were made at CRM 14.4, CRM 5.5, and at the Kingston Steam Plant. Also, a dye concentration survey in the channel of Emory River was made on Friday, August 30. Samples for radionuclide analysis were taken every two hours from the nappe of White Oak Lake outflow and every six hours and at times of peak dye concentrations at CRM 14.4 and CRM 5.5. At the Kingston Steam Plant a continuous sample of raw river water was taken from a tap on a condenser intake pipe.

*g/min = grams per minute.

Primary effort has been directed to an analysis of the data on dye concentrations, particularly at CRM 14.4 and CRM 5.5. As mentioned earlier, both the dye injection rate and the discharge from White Oak Lake were constant during the study period. The predicted values for these two stations (CRM 14.4 and 5.5) were calculated on the basis of estimated dispersion and dilution, and the observed values were compared with the predicted values. Figure 25 shows the comparison for CRM 14.4, and Fig. 26 for CRM 5.5. The maximum observed dye concentration at the Kingston Steam Plant was about 12.0 ppb at 2:45 p.m. on Wednesday, August 28, and the maximum dye concentration observed during the August 30 fluorometer traverse of Emory River was about 5 ppb at ERM 0.4.

A comparison of the predicted concentrations at CRM 14.4 (Fig. 25) and CRM 5.5 (Fig. 26) with the observed values shows that the predictions were reasonably good, even though the predicted concentrations, in general, were slightly higher than the observed concentrations. A better knowledge of the dispersion characteristics would enable more accurate predictions to be made, and some further field observations under "winter conditions" have been proposed. Further measurements will be necessary to define the minimum effective dilution in the Clinch River under the variety of conditions that will occur during power releases of water at Melton Hill Dam. On the basis of results from the dye tracer tests, the momentary dilution as affected by power releases will be about 10 times less than the median (see page 125). It was concluded tentatively that the radionuclide concentrations at Gallaher Bridge will remain below MPC_w during the summer months after power releases from Melton Hill Reservoir have begun.

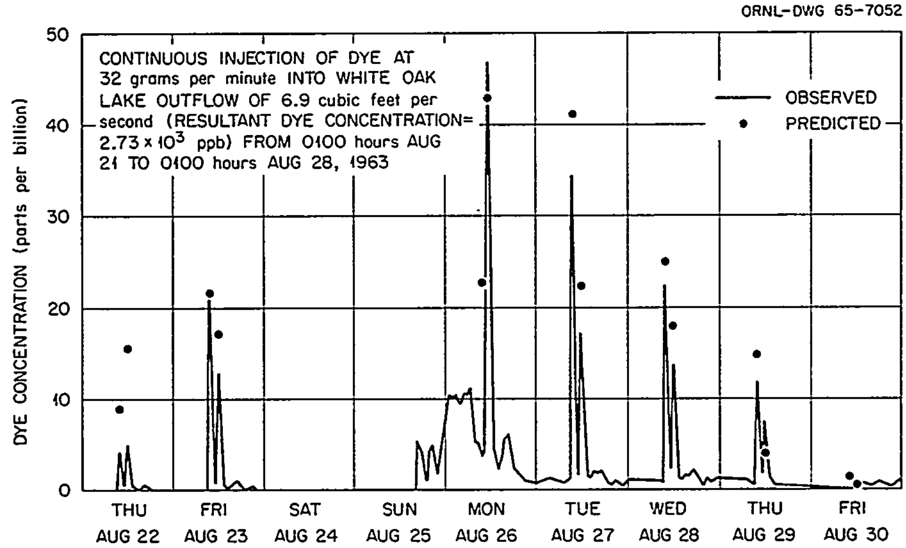

Fig. 25. Variation in Rhodamine B Concentration with Time During Period August 22–30, 1963, at Clinch River Mile 14.4

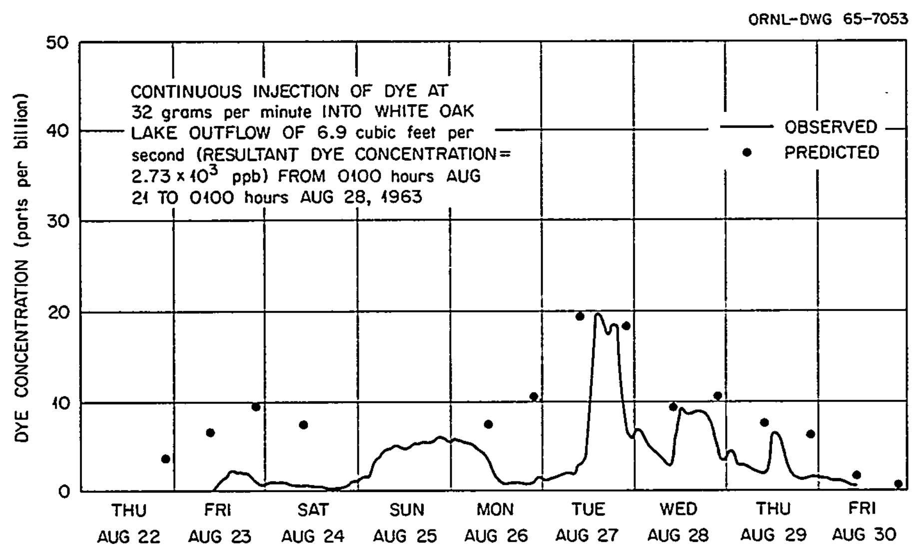

Fig. 26. Variation in Rhodamine B Concentration with Time During Period August 22–30, 1963, at Clinch River Mile 5.5

Temperature Observations. - In addition to the calculations and observations of dye concentrations, 24-hr observations of the movement of the temperature stratification line in Clinch River between CRM 4.5 (Centers Ferry) and CRM 23.1 (Melton Hill Dam) were made. These observations, made with the cooperation of TVA, were begun on Thursday, August 22, and continued on Sunday, August 25. Temperature profiles were made at about 5-mile intervals from CRM 20.8 downstream to the junction of Clinch River and Emory River and at 3-mile intervals from that point upstream in the Emory River to ERM 12 on August 21, 22, 25, 26, 27, and 28.

The temperature profile of the river during the 8,000-cfs flow showed that the temperature was uniform, 19°C, from CRM 23 to CRM 10. When the flow ceased on Friday afternoon, there was an unstable situation in the Clinch River, with 19°C denser water in the upper part of the river and warmer, lighter water, 29°C, downstream. Consequently, the heavier water drained out of the reach, and the lighter water flowed in until a more stable configuration was reached.[51] Later profiles showed that by 11:00 a.m. on Sunday, August 25, 22°C water had reached upstream to CRM 23. Therefore, 0.48 feet per second (fps) was the minimum velocity at which the heavier (colder) water could have drained out of the reach and the lighter water flowed upstream into the reach.

Theoretical Investigations--Calculations of Predicted Concentrations During the First and Second Dye Tests. - In order to make calculations and to predict concentrations and times of arrival in Clinch River of known releases of a dye or radioactive material at White Oak Dam, it was necessary to assume a normal pattern of discharge at Melton Hill

Dam and a steady outflow from White Oak Creek through White Oak Dam. A regular pattern of expected power releases at Melton Hill Dam is shown in Fig. 23. For purposes of these calculations, it was assumed that the steady flow in White Oak Creek through White Oak Dam was 7 cfs.

The ebb and flow of water in White Oak Creek embayment due to the power releases at Melton Hill Dam are much the same as flow in a tidal estuary. With techniques of tidal flow analysis, it was possible to compute the total mixing in the embayment and the net rate of movement through the embayment into the Clinch River. Also, it was possible to compute the predicted maximum concentration of the dye pulse as discharged from the creek and at downstream sections in the river and to estimate the travel times of the pulse to these sections. A method similar to the Woods Hole Analysis based on the "tidal-flushing" theory as advocated by Ketchum was adopted.[52] Variations of dye concentrations with time during the second test as predicted by this method are compared with the observed values at CRM 14.4 and at CRM 5.5 in Figs. 25 and 26 respectively. The basis for these calculations is summarized below. The methods of calculation were summarized in a report on the 1963 dye-tracer studies, presented to the Steering Committee on December 4, 1963.[49]

It was assumed that there would be: (1) complete mixing in the embayment during the tidal cycle; (2) no flow out of White Oak Creek into the river during releases from Melton Hill Dam; and (3) a constant outflow of 7 cfs from the creek through White Oak Dam into the embayment. The 0.6-mi-long reach of the White Oak Creek embayment was divided into 12 sections, all of equal volume, which in the calculations corresponded with the "volume-segment" subdivisions used by Ketchum in evaluation of an estuary.[52]

The embayment has an average cross section, when the river elevation (el) is 741 feet above MSL (mean sea level), of 265 sq ft with a volume of 6.3×10^6 gal. At el 745 ft the average cross section is 875 sq ft, and at el 750 the average cross section is 2360 sq ft, with proportionately greater volumes in the embayment. When the Melton Hill Dam releases 16,000 cfs, this causes a rise in the water surface elevation at the mouth of White Oak Creek of 2.8 ft to el 743.8. A release of 8,000 cfs causes a rise of 1.6 ft to el 742.6. These rises are equivalent to water-volume increases of 9.4×10^6 gal and 4.5×10^6 gal, respectively.

Using the above assumptions and available information on river elevations and embayment volumes, the pollutant (dye) could then be routed section-by-section through the embayment during successive tidal cycles, and the time and concentration of its flow into the river could be calculated. Assuming that each injection of dye into the Clinch River would remain together and function as a single unit, it was possible to route the mass of contaminant (the dye) downstream.

At 10 p.m. on Friday (August 23) the weekend shutdown of water releases occurred and continued for 58 hours until 8 a.m. the following Monday. Flow of water and dye from the White Oak Creek embayment into the still river water commenced soon after the shutdown began and continued until the startup of Melton Hill releases at 8 a.m. Monday morning. On Sunday afternoon, August 25, at about 3 p.m., aerial observations were made. It was observed that a continuous mass of dye extended from CRM 20.0 to CRM 22.5; the mouth of White Oak Creek is in this reach at CRM 20.8.

The effective dilution in the Clinch River of discharges through White Oak Dam is less during the conditions associated with power releases. The minimum effective dilution factor was about 50 during the test period of intermittent releases and a weekend shutdown at Melton Hill Dam. On the basis of the concentration of tracer injection at White Oak Dam and the peak concentration observed on Monday, August 26, the effective dilution at the Gallaher Bridge sampling site was reduced by a factor of about 10 as compared with a median dilution of 570 for the period 1950-1960.[53]

The twice-daily pulses of dye from the creek were observed on the week days during the test period between about 10 a.m. and 11:45 a.m. at the Gallaher Bridge observation station (CRM 14.4). During the weekend increased concentrations unexpectedly occurred at this station on Sunday afternoon (see Fig. 25). When there is no release from Melton Hill Reservoir, upstream flows might occur because of river-control operations in the Tennessee River, re-establishment of thermal stratification in the Clinch River, or as a result of a combination of both processes. These upstream flows, though minor, could move a mass of dye short distances. It is believed that the mass of dye that passed the Gallaher Bridge section at 11:45 a.m. on Friday of the first week and was stagnated about CRM 12, slowly moved upstream later during the weekend and finally stagnated in the vicinity of the sampling site beginning on Sunday afternoon.

It has been shown that reasonable prediction of the effects of a sequence of power releases from Melton Hill Reservoir could be made if the dispersion characteristics of the releases were empirically known.[50] A more general analytical method of predicting dispersion due to these power releases is being sought. One possible method of prediction is to assume that the factors which influence variations in the dispersion characteristics are the same for the unsteady nonuniform flow occurring in the Clinch River as for steady uniform flow. In steady flow no variation in flow or in velocity occurs with time. In uniform flow no change in velocity occurs with distance; that is to say, there is no convective acceleration either in the direction of flow or in the direction normal to flow.[54]

For the predictions of concentrations in the stream, the diffusion coefficients and also the energy gradients for the prevailing conditions must be determined or assumed. Data for evaluation of these two factors are presently very limited. In a waste treatment and disposal progress report [54] mention of the variables relevant to prediction of stream concentrations and a discussion of possible approaches and equations that may be applicable to determine the diffusion coefficient are given. Figure 27, taken from this progress report, shows the variation in the diffusion coefficient as computed for a number of points at which observations were made during tracer studies in August 1961 and February 1962. The variation of the diffusion coefficient for the same tests with velocity of flow, one of the relevant variables is shown in Fig. 28. The flows in the river during these studies were steady.

To provide a better basis for analysis of unsteady and nonuniform flows the authors of the report to the Steering Committee[49] have stated several conclusions and recommendations which are summarized below.

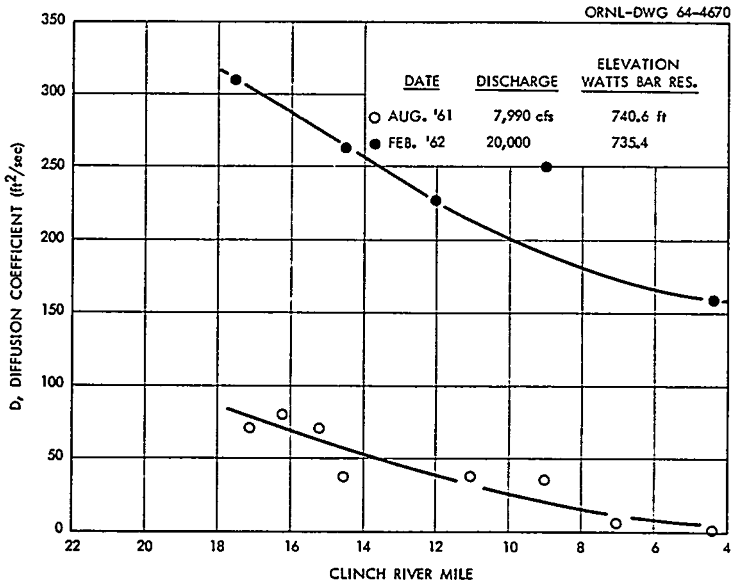

Figure 27. Longitudinal Variation in Diffusion Coefficient for Two Conditions of Steady Flow in the Clinch River

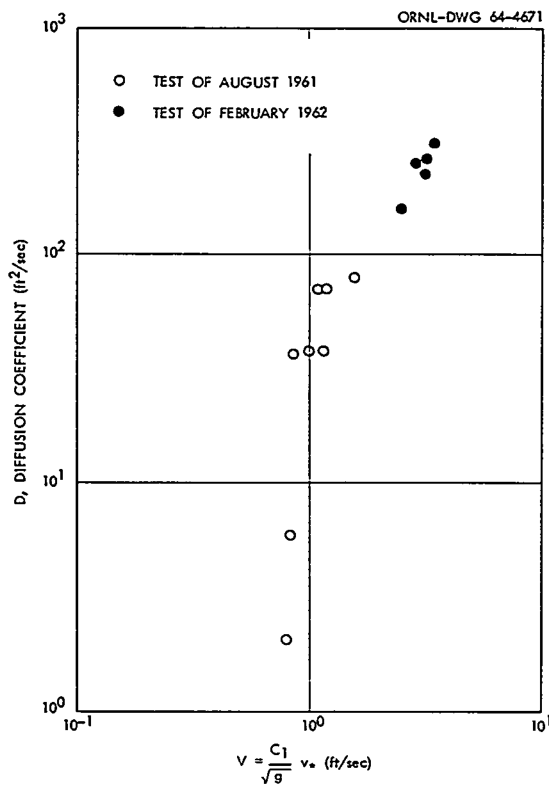

Figure 28. Relation of Diffusion Coefficient with Mean Velocity

Conclusions and Recommendations Regarding Further Field Studies of Dispersion in the Clinch River

Both local and general needs for better information are adequate justification for further field studies of the dispersion characteristics and behavior of discharges from White Oak Creek into the Clinch River, particularly studies under "winter" conditions and with unsteady and nonuniform flows.

Complete general analytical treatment of dispersion problems of the complexity encountered in the Clinch River will be very diffucult to achieve. In addition to difficulties in predicting the diffusion coefficients, mentioned above, there are uncertainties as to the degree of dispersion that will occur in the stilled pool of the Clinch River during periods of long shutdown such as prevailed on the weekend of August 24 and 25, 1963 during the second dye test.

Tests with tracers, as previously described, will indicate the general magnitude of dilution, rates of travel, and duration of appreciable levels of contaminants at critical locations for particular conditions. However, more exact determinations of the effects of power releases on the dispersion of radioactivity in the Clinch River for the variety of flow conditions might be better obtained by a materials balance study based on the input and output of contaminants in the study reach. This approach was recommended.

For the materials balance study the input load of radionuclides to the study reach could be determined at White Oak Dam and at Melton Hill Dam. A continuous proportional water sampling station is in operation at White Oak Dam and, also, in the tailrace of Melton Hill Dam. Analyses of samples from these two stations could be made to provide the input data required for a materials balance study.

On the basis of previous work during the Clinch River Study, the output load for the materials balance study might be determined at two locations, the intake to the water plant of ORGDP (CRM 14.4), and above Centers Ferry (at CRM 5.5). It was recommended that continuously operating proportional samplers be used at these sites. The development and fabrication of such equipment would cost about $5,000 for each station.

Because of the high cost, the construction of more than one downstream sampling station may not be feasible. From the standpoint of problems in the development, installation, and maintenance of one such station only, the preferred site is the one at CRM 14.4. General methods of actuating the equipment at either of the two downstream stations mentioned above were suggested in the report to the Steering Committee. It was recommended that, with these sampling facilities available, a system of coordinated sampling and analysis upstream and downstream from the mouth of White Oak Creek should be established to provide comparative data on radionuclide concentrations in the river. This should be carried on for several months selected to represent both summer and winter conditions after the Melton Hill Dam power station is in operation.

In the report it was concluded that, with the development of better information for determining diffusion coefficients and energy gradients for various conditions, much better predictions of dispersion and resultant concentrations of contaminants released to the river could be made. It was recommended that any opportunities for further field studies that would aid in the quantitative definition of these parameters should be considered, and followed up if possible.

SAFETY EVALUATION STUDIES

Analyses of the safety of conditions that result from discharges of radionuclides to the Clinch River and the potential exposure of people to ionizing radiation from these discharges have been made by the Subcommittee on Safety Evaluation. This subcommittee was established by the Steering Committee October 27, 1961, "to study available information and additional data that may be obtained and evaluate the potential hazards of discharges of radioactive wastes from Oak Ridge installations in the Clinch and Tennessee Rivers."[55]

The work of this subcommittee and some of the results obtained were summarized in Status Report No. 4 on the Clinch River Study.[56] In this summary the bases of safety evaluation, the objectives of the subcommittee, an outline of additional information needed, data on the radionuclides released and concentrations in the rivers, the avenues of human exposure, the critical organs considered, and the radioactivity in the water of the Clinch and Tennessee Rivers in terms of MPC_w were reviewed. Also, the estimated potential radiation dosages from drinking water, water immersion, and radioactivity in bottom sediments were given.

The summary in Status Report No. 4[56] was based on a preliminary safety analysis prepared by the subcommittee in 1963. Since that time the subcommittee has completed its evaluation and submitted to the Steering Committee its final report which contains the revised version of the entire safety analysis.[57] In addition to the explanatory sections and estimates of dosages outlined above the subcommittee's final report includes calculated estimates of radiation dosages for various populations which might result from drinking water from the Clinch or Tennessee River, eating contaminated fish, or eating food from crops irrigated with contaminated river water.

Regarding the limitations of its analysis, the subcommittee recognized that any biological form may receive some degree of radiation exposure but this study confined its efforts to calculation of radiation dosages to man. Since data on direct measurements of dosages were not available and could not be obtained, the exposures were calculated from determinations of the amounts of radioactive material in the various environmental media and by the use of assuptions as to the fraction of this material that might cause internal exposure of the affected population. Not all of the desired information was available. For example, in order to complete some calculations it was necessary to estimate the dietary habits and amounts of principal food stuffs consumed as well as occupational and recreational patterns. Several estimates of external radiation exposures were calculated from measured concentrations of radionuclides in environmental media but these should be confirmed by direct measurements where possible.

The tabulated estimates of exposure values from various media derived by the subcommittee will not be included in the present report because: the preliminary analysis was described and partially summarized in Status Report No. 4;[56] and (2) the final report of the subcommittee is being prepared for release as a supplement to the present report (Status Report No. 5) and should become available about the same time.[57]

As a summary of its final report the subcommittee included at the end a section on conclusions and recommendations which were based on the detailed data in the earlier sections of the report. The comments below are based mainly upon the subcommittee's conclusions, particularly those regarding the estimates that were not summarized in Status Report No. 4.[56]

Estimates of Radiation Dosages

On the basis of concentrations of radionuclides in the various environmental media, and using the approach and assumptions described in the final report,[57] the subcommittee calculated the potential dosages to the skeleton, total body, and thyroid of man due to consumption of contaminated water and fish. These were found to be the critical exposure possibilities but estimates were also made of external radiation dosages from water immersion or contaminated sediments in the rivers, and from radionuclide concentrations in water treatment plants and systems. A hypothetical analysis of possible future hazards that might develop from irrigation of food crops was made although the present practice of irrigation is limited.

As a basis for interpretation of the potential effects from the calculated exposures from the individual vectors and the aggregate overall exposure from all of the vectors the subcommittee used the criteria of the International Commission on Radiological Protection (ICRP),[58] and the Radiation Protection Guides (RPG) recommended by the Federal Radiation Council (FRC) for normal peacetime operations.[59] The recommended limits from reports by ICRP and FRC were compared by the subcommittee in its report.[57] It was shown that the differences were minor and within the precision of the data employed by these agencies in arriving at the suggested limits; and that differences in the acceptable rates of intake were not significant.

In its analysis of the potential hazard of internal dose from ingestion of water or contaminated fish the subcommittee concluded that ^{90}Sr has been the most important of the critical radionuclides released to the Clinch River, contributing more than 99% of the dose to the skeleton and total body and 70% of the thyroid dose. However, it was noted that in the recent Publication 6 of ICRP[60] the recommended permissible intake of ^{90}Sr by standard man was increased because it was considered that the metabolic data provided a better estimate of acceptable values for ^{90}Sr (in bone as the critical organ) than the single exponential model used previously. This change will result in an overestimate in this report of the potential exposures from ^{90}Sr to the total body and the skeleton by a factor of about 2 and 4 respectively. It was estimated that the total dose to the skeleton of the critical population was smaller by a factor of about 5 than the allowable dose from contaminated drinking water; improved waste management at ORNL has resulted in a decrease in ^{90}Sr released to the Clinch River.

A table in the report gives the estimated cumulative dose received by critical organs of males from use of Clinch and Tennessee Rivers for drinking water, recreation, and production of food fish and, for comparison, a statement of the recommended maximum permissible dose to the skeleton and total body is given. In commenting on this table the subcommittee again noted that: (1) the calculated internal dose from ingestion of drinking water and contaminated fish were the critical exposure pathways; (2) immersion in water, contact with bottom sediments, and occupational work within a water treatment plant were not significant from the standpoint of comparative exposure; and (3) the total cumulative dosages received by the critical organs have been well within the limits recommended by ICRP, either for occupational exposure or for the general population.

The report also mentions the several assumptions upon which the calculations were based and the consequent limitations in use of the data. Because of the inadequacy of quantitative information presently available and the conservative assumptions, it is apparent that the estimated dosages would be less if better information were available and were factored into the calculations. One excellent example is the recent change in the MPC_w for ^{90}Sr mentioned above which would reduce the estimated dose to the skeleton by a factor of 4 and to the total body by a factor of 2.

Crop Irrigation

The subcommittee noted and gave references to show that irrigation of a variety of crops has been practiced in Tennessee, for at least 50 years, and that during the period 1950-1955 irrigation with water from the Tennessee River was significantly increased. From a survey in 1958 it appeared that of 1,021 irrigation units in operation at that time, 20% were used for irrigating truck crops and 30% for irrigating feed crops (corn, silage, and hay). In West Tennessee, west of the Tennessee River, ground water is the principal source of water for irrigation but surface water is predominantly used in East Tennessee. More than 15 inches of water have been applied to truck crops during the growing season.

Currently crop irrigation along the Clinch River is nonexistent and there is little practice of irrigation along the Tennessee River downstream from Oak Ridge. Therefore, a safety analysis of the possible transfer of fission products from contaminated river water to foods by crop irrigation is largely a hypothetical exercise. It is considered justifiable, however, in order to point out future-problem areas and, where possible, to uncover long-term problems that may result from currently unpredictable usage of this natural resource. The subcommittee made suitable conservative assumptions and calculated potential exposures in spite of the lack of specific data regarding such vital factors as: types of crops irrigated, quantity of water applied per year, percentage of total production that might be affected by irrigation, the concentrations of particular radionuclides that might be accumulated in the soil, the factors of plant uptake of radioactive elements from the soil, the concentrations of radioactive materials that might be found in foodstuffs, and the proportion of food crops irrigated with river water that might be used by the average population as compared with unirrigated food crops. The probable present and the potential future radiation dosage to the population that might be attained from the use of irrigated crops was estimated.

In its interpretation of these estimates the subcommittee concluded that at the present time there is no danger of significant quantities of fission products entering man's diet because of crop irrigation. Truck crops in this environment contribute little to the total quantity of produce and are grown only on small plots and for a short period during the year. In areas where climatic conditions are more conducive to year-around irrigation of large agricultural plots, however, crop irrigation with contaminated water could take on greater importance. Some use can be made of data provided by fallout studies but there is need for data from experiments that will define radionuclide entry into man's diet from contaminated irrigation water. Analyses of samples of soils and crops that are irrigated with Tennessee River water, although difficult, might assist in defining the extent of fission product concentrations due to such practice.

Some Comments on the Subcommittee's Conclusions

From the four pages of conclusions and recommendations to be published in the subcommittee report,[57] the following are selected for brief comment:

1. Disposal of radioactive wastes to the Clinch River has resulted in radionuclide concentrations in the river water below FRC recommendations. Most important as a potential source of radiation exposure was consumption of contaminated water and fish, but no population groups subjected to these critical exposure pathways received excessive dosages. If the practice of irrigation with river water continues to develop, the long-term effects of crop irrigation with the contaminated water could become the most important avenue of exposure.

2. The calculation of internal dose to critical organs should be revised using the actual concentrations of radionuclides in the river water as determined by the Subcommittee on Water Sampling and Analysis. Internal dose calculations based solely on exposure of so-called standard man would underestimate the dose likely to be received by critical population groups (the most highly exposed groups) by a factor of at least 2. Improvements in dose estimates are possible if better information can be obtained. For example, when information becomes available concerning the differences in metabolic rates and processes in children and adults as they relate to the important radionuclides it can be factored into the mathematical models (given in the report) and the dose to the critical organs recalculated.

3. Strontium-90 has been by far the most important of the critical radionuclides in the liquid wastes released to the Clinch River. As mentioned earlier, it has

contributed more than 99% of the dose to the skeleton and the total body and 70% of the thyroid dose. Ruthenium-106, ^{137}Cs, and ^{60}Co contributed significantly to the dose received by the GI Tract. The doses are well below and often are only a small percentage of the permissible dosage limits. The actual values of the estimated dosages received are given in the report.[57]

4. Routine environmental monitoring should emphasize mainly the critical pathways of human exposure which currently are the consumption of contaminated water and fish. Periodic review is needed to confirm the adequacy of the monitoring program and assure that exposures from radionuclides of short half life as well as of long half life are considered. Minor sources of exposures such as contaminated water and bottom sediments should be monitored, although less intensively, in order to be sure that the over-all potential of human exposure is assessed periodically.

5. There is need for continued research studies to investigate radionuclide transfer to fish and to irrigated crops. The types of data needed have been summarized earlier. A potentially important source of entry of ^{90}Sr into man's food chain can be eliminated if fish can be prevented from inhabiting White Oak Creek or White Oak Lake and then entering the Clinch River. An adequate analysis of crop irrigation as a critical exposure pathway requires knowledge of fission product behavior in soils and plants as well as of the extent and methods of irrigation.

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CONCENTRATIONS, TOTAL STREAM LOADS, AND MASS TRANSPORT OF RADIONUCLIDES IN THE CLINCH AND TENNESSEE RIVERS

Supplement No. 1 to Status Report No. 5
on Clinch River Study

M. A. Churchill
J. S. Cragwall, Jr.
R. W. Andrew
S. Leary Jones

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HEALTH PHYSICS DIVISION

CONCENTRATIONS, TOTAL STREAM LOADS, AND MASS TRANSPORT OF RADIONUCLIDES IN THE CLINCH AND TENNESSEE RIVERS

Supplement No. 1 to Status Report No. 5 on Clinch River Study

By

M. A. Churchill
J. S. Cragwall, Jr.
R. W. Andrew
S. Leary Jones

Progress Report No. 4 (final) of Subcommittee on Water Sampling and Analysis. Presented at Meeting of Clinch River Study Steering Committee, December 15-16, 1964

AUGUST 1965

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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PREFACE

The Subcommittee on Water Sampling and Analysis was established on September 22-23, 1960, by the appointment of the following members: M. A. Churchill (TVA), Chairman, J. S. Cragwall (USGS), A. G. Friend (USPHS), and S. L. Jones (TDPH). Following the transfer by the USPHS of Dr. Friend to Boston, R. W. Andrew (USPHS) was appointed to replace him.

Although this report includes a statement of the objectives of the Subcommittee, the method of study, detailed findings, and recommendations, some of the details of procedures used are omitted here since these have been included in previous Status Reports. Details of water sampling procedures at the established sampling stations are given in Status Report No. 1, pp. 22-23, and p. 73, Status Report No. 2, pp. 8-15, Status Report No. 3, pp. 10-11, and in Status Report No. 5, p. 16. Information concerning procedures used for radiological determinations is given in Status Report No. 1, p. 23, Status Report No. 2, pp. 16-17, Status Report No. 3, p. 12, and in Status Report No. 5, p. 16. (See references 2, 3, and 4, p 72.)

Results of stable chemical determinations for the sampling period November 1960-March 1961 are included in Status Report No. 2, pp. 29-33, and results for the period April 1961-June 1961 are included in Status Report No. 3, pp. 30-32. Analytical results on samples collected in the period July 1961 through November 1962 are available in Progress Report No. 3, Subcommittee on Water Sampling and Analysis, February 6, 1963. In addition, a statistical analysis of all the stable chemical data is included in Status Report No. 5, pp. 18-21. (See references 2, 3, and 4, p 72.)

Acknowledgements

Special acknowledgment is due R. W. Andrew for his initiative and painstaking thoroughness in re-examining and correcting the electronic computer program used to determine concentrations of three of the four radionuclides of primary interest. Acknowledgment is also due R. A. Buckingham formerly of TVA, for his valued assistance in preparing Subcommittee Progress Reports Nos. 1, 2, and 3. Similar acknowledgment is due W. R. Nicholas, TVA, for his assistance in preparing Progress Report No. 4 of the Subcommittee, which report, essentially unchanged, is reproduced here.

CONTENTS

ILLUSTRATIONS

PROGRESS REPORT NO. 4 (FINAL)

Subcommittee on Water Sampling and Analysis

Clinch River Study

December 15-16, 1964

Purpose of Work

The basic purpose of the work of the Subcommittee on Water Sampling and Analysis is to collect and interpret such information concerning radionuclides suspended and/or dissolved in the waters of the surface streams downstream from Oak Ridge, Tennessee, as will assist the Clinch River Study Steering Committee in pursuing the basic purposes of the entire study, namely, "(1) to determine the fate of radioactive materials currently being discharged to the Clinch River, (2) to determine and understand the mechanisms of dispersion of radionuclides released to the river, (3) to evaluate the direct and indirect hazards of current disposal practices in the river, (4) to evaluate the over-all usefulness of the river for radioactive waste disposal purposes, and (5) to recommend long-term monitoring procedures."

Included as part of the work of the subcommittee is the determination of the mineral quality of the surface waters involved in the over-all study.

Method of Study

General--The general plan of the study involved systematic collection and analysis of water samples at selected sampling stations. Daily subsamples of water, the individual volumes of which at each station (except at Loudon) were proportioned to the volumes of daily streamflow passing that particular station, were composited weekly for analysis (monthly for most mineral analyses). Such analyses provided the mean flow-proportioned concentration of each radionuclide of interest passing each station each week. By combining this mean concentration with the total flow of water passing the station during each week, the total load, in curies, of each radionuclide passing the station was determined. The cumulative load of each radionuclide at each station was plotted progressively with time. The mass curves so produced reveal on comparison, one with another, the quantitative loss (by sedimentation, biological uptake, etc.) or gain (from fallout on the watershed) of this particular radionuclide between successive downstream stations.

The Centers Ferry sampler malfunctioned (See Progress Report No. 3, of the Subcommittee on Water Sampling and Analysis, page 27) during September through November 1961. This malfunction possibly affected the suspended-sediment results for all radionuclides. The degree to which the results are affected is dependent upon the proportion of a given radionuclide associated with the suspended solids.

Sampling Stations--Sampling stations used in the study are located as follows, and as shown in figure 1:

1. Clinch River at Oak Ridge water plant--Clinch River mile 41.5
2. White Oak Creek at White Oak Dam, mile 0.6
3. Clinch River at Gallaher Bridge--Clinch River mile 14.6
4. Clinch River above Centers Ferry--Clinch River mile 5.5
5. Tennessee River at Loudon, Tennessee--Tennessee River mile 591.8
6. Tennessee River at Watts Bar Dam--Tennessee River mile 529.9
7. Tennessee River at Chickamauga Dam--Tennessee River mile 471.0

Period of Sampling--Except for the station at Gallaher Bridge, sampling was begun in November 1960 and extended through November 1962. At Gallaher Bridge, sampling was begun on January 8, 1962, and was discontinued at the end of November 1962.

Sampling Procedures--Sampling procedures at each sampling station have been explained in detail in previous progress reports.

Radiological Determinations--The radionuclides of primary importance in the Clinch River Study, in the order named, are strontium-90, cesium-137, cobalt-60, and ruthenium-106. Consequently, determinations were made of concentrations and total loads of these radionuclides. All radiological determinations in this study have been made by the U. S. Public Health Service, Cincinnati, Ohio.

Details of sample preparation and analysis have been explained in previous progress reports.

Stream Discharges--The necessary data on streamflows at the five upstream sampling stations have been provided by the U. S. Geological Survey, through the cooperation of its Tennessee District. Discharges at Watts Bar and Chickamauga Dam have been supplied by TVA. (See plotted streamflow data in figures 2 and 3.)

Mineral Analyses--All mineral analyses were made in Nashville, Tennessee, by the staff of the Tennessee Stream Pollution Control Board. Methods used and results obtained have been included in previous progress reports.

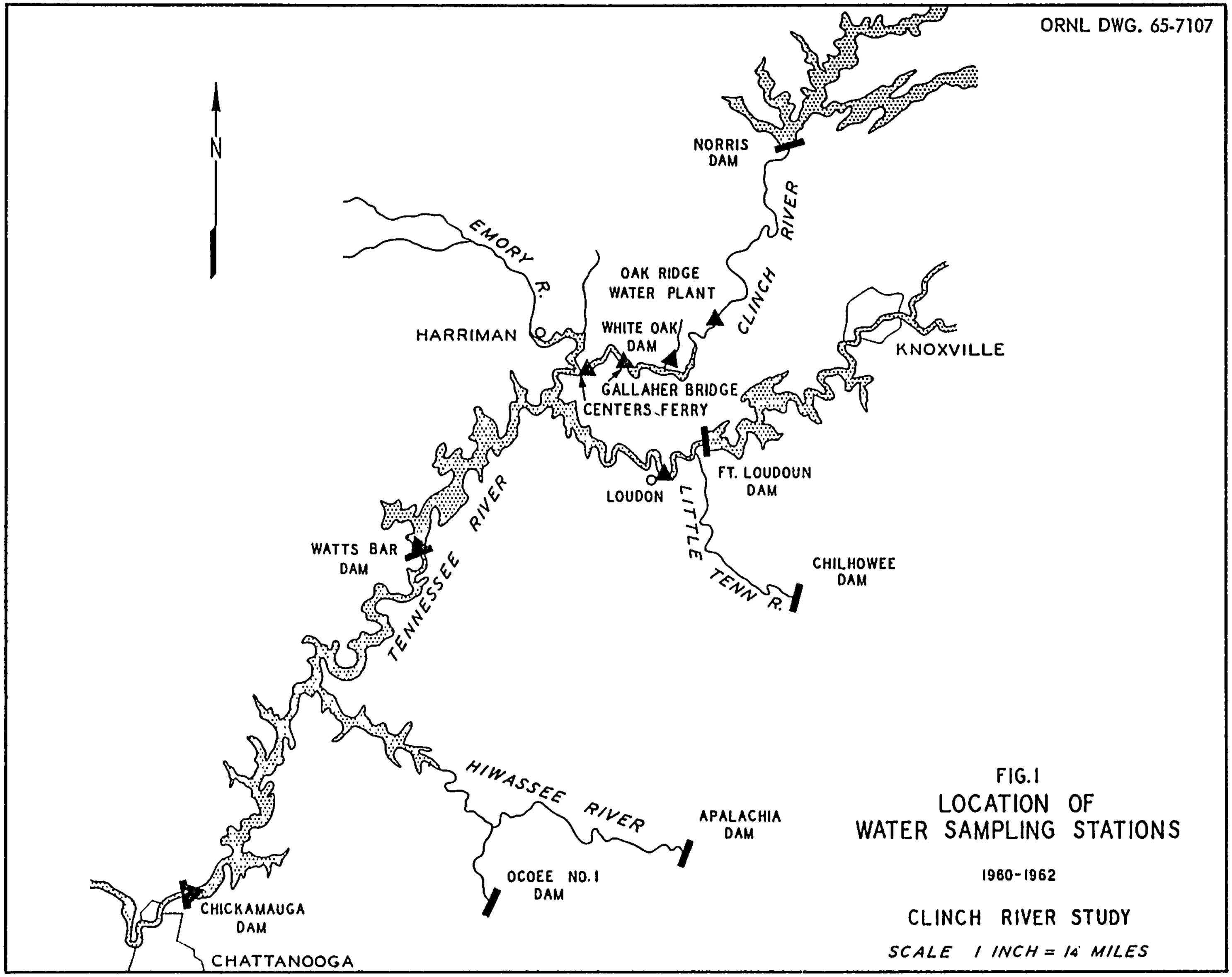

FIG. 1
LOCATION OF
WATER SAMPLING STATIONS

1960-1962

CLINCH RIVER STUDY

SCALE 1 INCH = 14 MILES

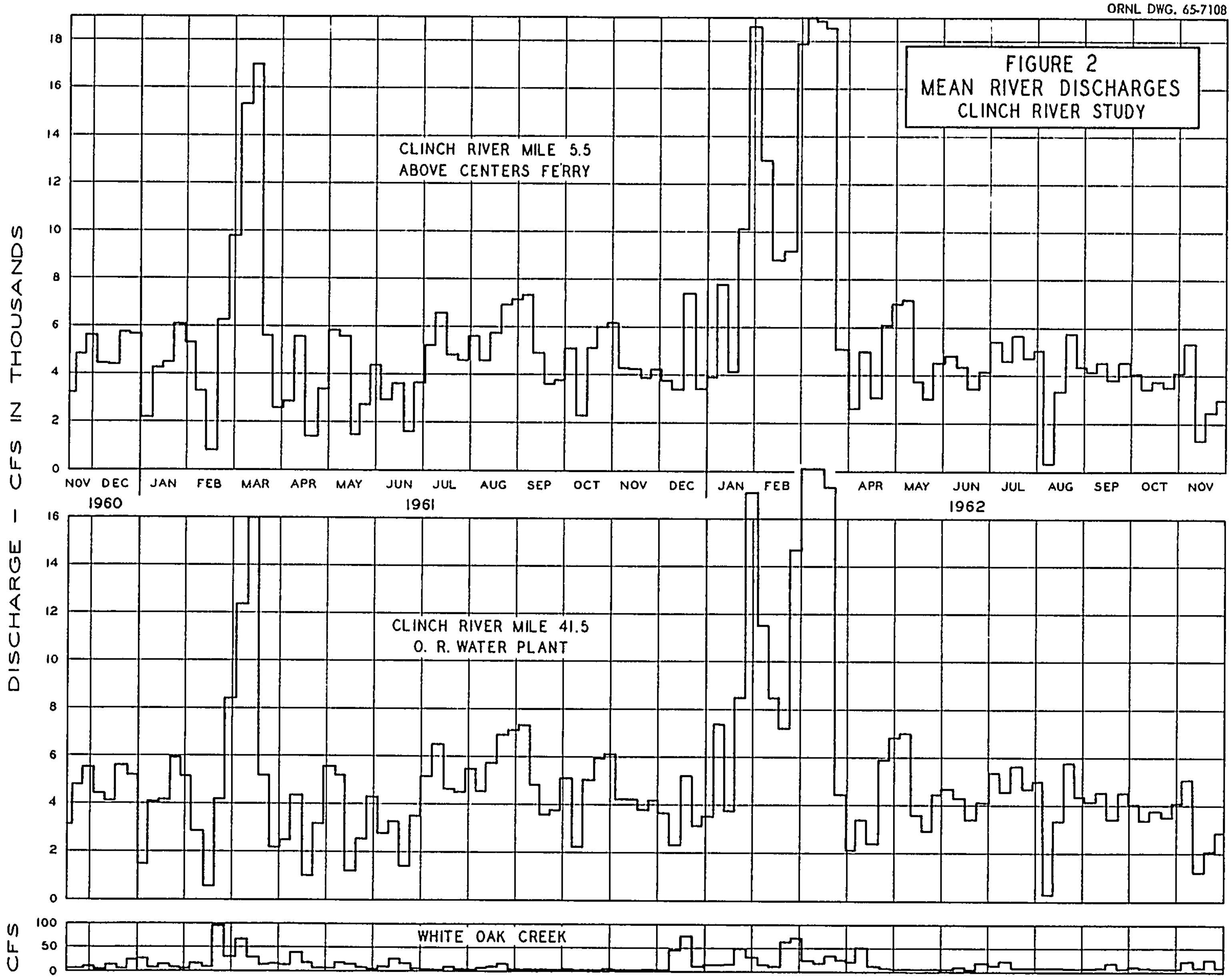

FIGURE 2
MEAN RIVER DISCHARGES
CLINCH RIVER STUDY

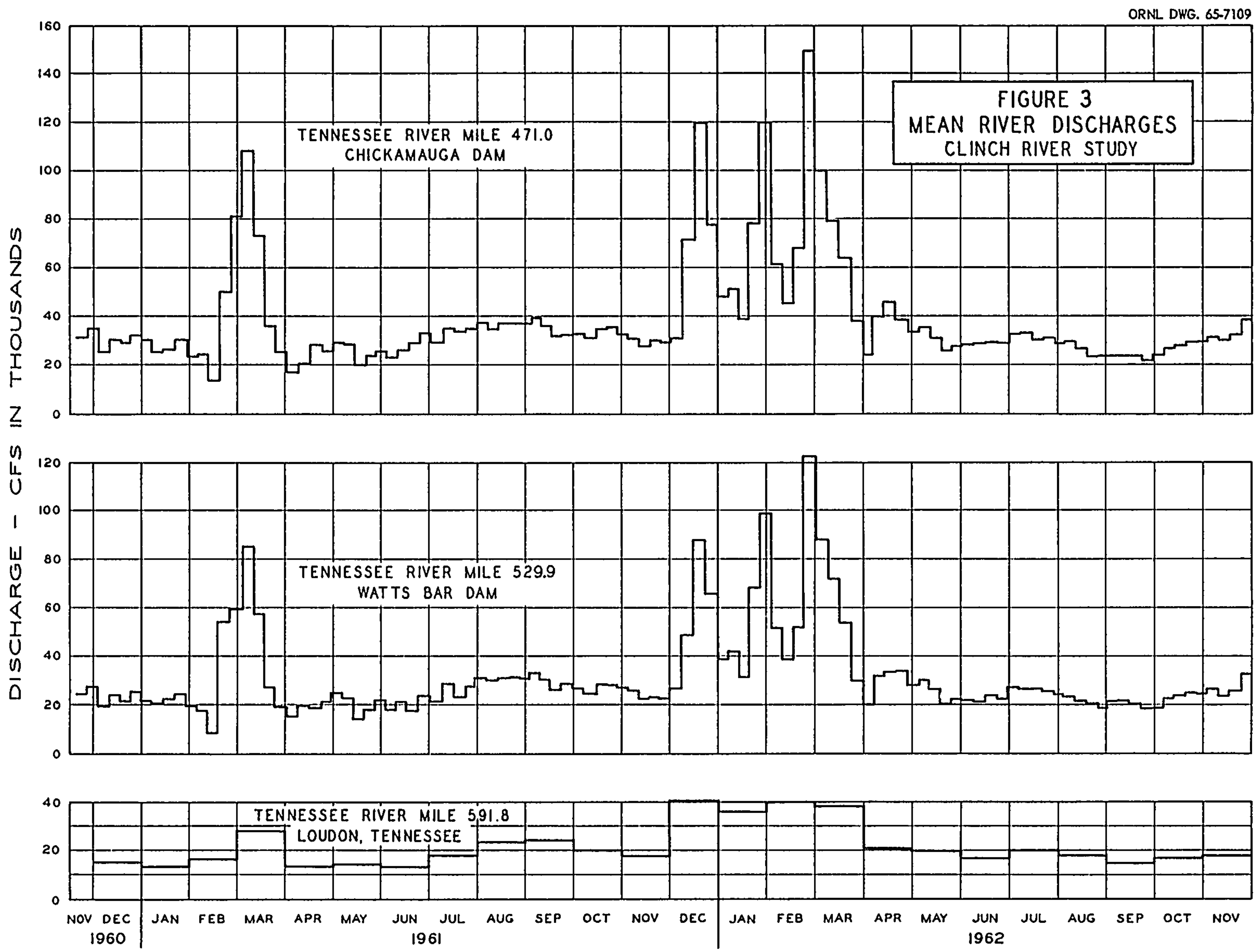
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FIGURE 3
MEAN RIVER DISCHARGES
CLINCH RIVER STUDY
TENNESSEE RIVER MILE 471.0
CHICKAMAUGA DAM
TENNESSEE RIVER MILE 529.9
WATTS BAR DAM
TENNESSEE RIVER MILE 591.8
LOUDON, TENNESSEE
DISCHARGE – CFS IN THOUSANDS
160
140
120
100
80
60
40
20
0
NOV DEC JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV
1960
1961
1962

Revision and Extension of Data Previously Reported

Progress Report No. 3, issued February 6, 1963, reported results on the four radionuclides of interest to this study from the beginning of sampling in November 1960, well into the summer of 1962. However, due primarily to a reexamination and corrections made by personnel of the U. S. Public Health Service of the electronic computer program used to determine concentrations of cesium-137, cobalt-60, and ruthenium-106, many major changes were made in previously reported concentrations of these three radionuclides. The program in error systematically produced results that were 50 percent to 100 percent too high, for the samples of larger size (i.e., all samples except those for White Oak Creek). Correction and updating of the computer program produced the results reported here. In addition, a few changes have also been made in previously reported concentrations of strontium-90 as a result of further checking of the sample calculations. Consequently, this report includes tabulated and plotted data on all four radionuclides that supersede the data reported in Progress Report No. 3. In addition, the data on all four radionuclides at all seven stations have been extended through November 1962, i.e., to the end of the two-year sampling period. The tabulated radionuclide results are accurate to no more than two significant figures. The additional figures were tabulated for statistical reasons only.

All data reported as negative values were assumed to be zero when determining loads for the mass diagrams. This probably gives a slight positive bias to the results.

Lower Limits of Detection of Radionuclides

To assist in judging the reliability of the mass curves, information concerning the lower limits of detection of radionuclides was obtained from the U. S. Public Health Service. These data are shown in the following table. Some values below these lower limits of detection are reported in the tabulation of radionuclide concentrations and were used for calculating the cumulative loads shown in the mass diagrams.

Approximate Lower Limits of Detection*
Picocuries per liter

	Strontium-90	Cesium-137	Cobalt-60	Ruthenium-106
White Oak Creek (TS and SS)	1	11	9	45
White Oak Creek (DS)	1	67	44	190
Other Samples (TS and SS)	0.03	1	1	2
Other Samples (DS)	0.03	4	2	11

*Estimated on the basis of 2 sigma counting error associated with a blank determination (background only). The presence of any other radionuclide in a given sample would tend to raise slightly the lower limits for cesium-137, cobalt-60, and ruthenium-106.

Strontium-90, Concentrations and Total Stream Loads

Concentrations of strontium-90 found in all samples at all stations for the two-year sampling period (strontium-90 data ended November 10, 1962, at all stations except White Oak Dam and Loudon) are shown in table 1. Since some counting error is probable for every sample, the true activity level (as determined by counting) in the sample is thought to fall within the range indicated by the magnitude of the plus or minus value (95 percent confidence limits) included with each reported concentration. The plus and minus values infer the level of precision in counting rather than the accuracy of the result since some additional uncertainty arises as a consequence of the chemical separation processes involved.

Maximum concentrations found in the weekly (monthly at Loudon) composite samples (including both suspended and dissolved solids) are shown in the following tabulation:

Maximum and Mean Concentrations of Strontium-90

Station	Highest Concentration pc per liter	Period of Occurrence	Flow-Weighted Mean Concentration pc per liter
Clinch R. at Oak Ridge water plant	5.0	12/4-10/60	0.71
White Oak Creek at White Oak Dam	17,450	11/13-19/60	1,349
Clinch R. at Gallaher Bridge	11.67	4/29-5/5/62	4.5*
Clinch R. at Centers Ferry	42.6	12/25-31/60	4.2
Tennessee R. at Loudon, Tenn.	2.3	January 1961	**
Tennessee R. at Watts Bar Dam	16.4	12/25-31/60	1.6
Tennessee R. at Chickamauga Dam	14.1	1/15-21/61	1.6

*Record begun January 8, 1962
**Not applicable.

In this tabulation, the values in the last column were obtained by dividing the total cumulative stream load for the period of record by the corresponding total volume of streamflow. The mean concentration so obtained is not the same as the mean concentration over time. (In fact, it is impossible to determine the mean concentration on a time basis from the basic data.) At only the White Oak Creek station does the flow-weighted mean concentration exceed MPC values for drinking water.

Table 1

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date	Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at		
					Loudon, Tenn.	Watts Bar Dam	Chickamauga Dam
1960							
11/13-19	TS 1.2 ±0.03	17,450 ±450		21.6 ±0.57			
11/20-26	TS 0.92±0.2	75.6 ± 3.9		7.5 ±0.2		6.7*	1.5 ±0.08
11/27-12/3	TS 0.2 ±0.1	640 ± 6.6		14.3 ±0.4		4.8 ±0.18	0.9 ±0.02
12/4-10	TS 5.0 ±0.4	4,770 ± 20		5.0 ±0.33	1.4* for	6.7 ±0.4	4.3 ±0.14
12/11-17	TS 3.1 ±0.3	1,730 ± 15		24.1 ±0.3	December	0.7 ±0.04	5.9 ±0.57
12/18-24	TS 0.5 ±0.1	6,280 ± 22		1.5 ±0.1		2.1 ±0.1	3.6 ±0.25
12/25-31	TS 0.62±0.09	7,070 ± 74		42.6 ±1.6		16.4 ±1.30	1.4 ±0.1
1961							
1/1-7	TS 2.0 ±0.2	878 ± 8.8		13.3 ±0.32		1.98±0.009	1.7 ±0.16
1/8-14	TS 0.2 ±0.03	15,900 ± 26.2		6.3 ±0.2		4.8 ±0.3	5.6 ±0.9
1/15-21	TS 1.9 ±0.1	2,875 ± 13		4.6 ±0.1	2.3 ±0.1	12.0 ±0.6	14.1 ±0.4
1/22-28	TS 0.5 ±0.1	2,032 ± 14		3.88±0.2	for January	5.1 ±0.24	2.4 ±0.2
1/29-2/4	TS 0.3 ±0.04	6,700 ± 62		9.9 ±0.37		2.7 ±0.2	3.75±0.23
2/5-11	TS 0.8 ±0.1	3,400 ± 25		37.0 ±0.6		1.2*	2.0 ±0.25
2/12-18	TS 0.29±0.04	6,600 ± 53		30*	0.39±0.07	0.3 ±0.1	1.3 ±0.1
2/19-25	TS 0.6 ±0.1	1,350 ± 10		11.9 ±0.2	for February	0.6 ±0.1	1.7 ±0.05
2/26-3/4	TS 0.4 ±0.02	1,060 ± 8		4.1 ±0.1		2.2 ±0.2	2.8 ±0.2
3/5-11	TS 0.3 ±0.1	590 ± 7		2.9 ±0.1		1.0 ±0.04	1.5 ±0.1
3/12-18	TS 0.33±0.06	160 ± 5		1.8 ±0.1	SS Bkgd.**	0.9 ±0.1	0.8 ±0.1
3/19-25	TS 0.3 ±0.02	930 ± 80		2.5 ±0.1	DS 0.3 ±0.04	2.0 ±0.2	1.0 ±0.1
3/26-4/1	TS 0.2 ±0.02	1,000 ± 80		2.3 ±0.2	for March	0.9 ±0.1	0.3 ±0.1

*Value is estimated.

**Bkgd. indicates background.

Blank spaces indicate data not available.

TS = total solids; SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1961	Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
4/2-8	TS 0.6 ±0.1	1,020 ± 25		5.3 ±0.1		0.7 ±0.05	1.3 ±0.1
4/9-15	TS 0.3 ±0.04	953 ±128		2.7 ±0.06	SS 0.05±0.009	1.5 ±0.2	1.5 ±0.07
4/16-22	TS 0.3 ±0.02	1,208 ± 50		4.7 ±0.08	DS 0.4 ±0.04	1.2 ±0.01	1.0 ±0.07
4/23-29	SS Bkgd.*	59.8 ± 0.09		0.1 ±0.01	for April	TS 1.3 ±0.09	TS 1.2 ±0.06
	DS 0.3 ±0.03	1,175 ±126		8.4 ±0.1			
4/30-5/6	SS Bkgd.	110.5 ± 1.2		0.1 ±0.01		Bkgd.	Bkgd.
	DS 0.3 ±0.03	2,225 ±130		2.8 ±0.07		0.8 ±0.04	1.2 ±0.1
5/7-13	SS Bkgd.	25.4 ± 0.8		0.1 ±0.01		0.1 ±0.01	0.1 ±0.1
	DS 0.3 ±0.03	1,462 ± 6		0.6 ±0.07		1.4 ±0.04	0.5 ±0.03
5/14-20	SS 0.1 ±0.02	18.8 ± 0.6		0.1 ±0.06	Bkgd.	Bkgd.	Bkgd.
	DS 0.3 ±0.02	1,400 ±100		4.5 ±0.1	0.3 ±0.03	1.4 ±0.03	1.6 ±0.07
5/21-27	SS Bkgd.	12.9 ± 0.5		0.1 ±0.01	for May	Bkgd.	Bkgd.
	DS 0.3 ±0.02	1,500 ±100		9.2 ±0.1		0.69±0.04	0.8 ±0.04
5/28-6/3	SS Bkgd.	30.0 ± 0.08		Bkgd.		Bkgd.	Bkgd.
	DS 0.5 ±0.03	2,000 ± 70		4.0 ±0.06		1.4 ±0.04	0.8 ±0.03
6/4-10	SS 0.1 ±0.01	71.8 ± 1.2		0.2 ±0.01		0.1 ±0.01	Bkgd.
	DS 0.16±0.03	1,400 ± 50		7.3 ±0.15		1.6 ±0.05	0.8 ±0.03
6/11-17	SS 0.2 ±0.02	130 ± 1.5		0.2 ±0.01	0.1 ±0.02	TS 1.80**	Bkgd.
	DS 0.4 ±0.03	1,800 ± 70		6.6 ±0.07	0.2 ±0.03		4.4 ±0.14
6/18-24	SS 1.23±0.07	80.2 ± 1.2		0.3 ±0.03	for June	Bkgd.	Bkgd.
	DS 0.3 ±0.02	1,679 ± 13		6.32±0.24		1.7 ±0.05	1.1 ±0.03

*Bkgd. indicates background.
**Value is estimated.

Blank spaces indicate data not available.
TS = total solids; SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
6/25-7/1	SS	Bkgd.*	50.8 ± 1.0		0.3 ±0.03		Bkgd.	Bkgd.
	DS	0.2 ±0.02	1,627 ± 35.4		5.0 ±0.08		1.5**	1.5 ±0.07
7/2-8	SS	0.2 ±0.02	78.1 ± 1.1		2.5 ±0.04		Bkgd.	Bkgd.
	DS	0.2 ±0.02	2,280 ± 43		3.0 ±0.06		1.2 ±0.06	2.2 ±0.06
7/9-15	SS	0.2 ±0.02	218 ± 2		Bkgd.	0.2 ±0.05	0.4 ±0.02	Bkgd.
	DS	0.3 ±0.02	2,565 ± 40.6		1.3 ±0.04	1.96±0.1	2.0 ±0.06	0.8 ±0.04
7/16-22	SS	Bkgd.	50.3 ± 0.09		0.8 ±0.07	for July	Bkgd.	0.2 ±0.01
	DS	0.8 ±0.05	2,195 ± 53		3.0 ±0.1		1.6 ±0.04	2.0 ±0.07
7/23-29	SS	0.4 ±0.05	27.7 ± 1.8		0.21±0.06		Bkgd.	0.05**
	DS	0.36±0.04	1,920 ± 50		3.07±0.18		1.7 ±0.05	1.29±0.06
7/30-8/5	SS	0.11±0.03	23.6 ± 1.3		0.06±0.02		0.6 ±0.01	0.01±0.02
	DS	0.19±0.02	1,560 ± 36		2.1 ±0.07		0.8 ±0.05	0.49±0.05
8/6-12	SS	0.15±0.03	39.0 ± 2.7		0.13±0.12	0.02±0.01	0.04±0.02	Bkgd.
	DS	0.07±0.02	2,025 ± 41		0.6 ±0.04	0.2 ±0.02	1.2 ±0.07	1.9 ±0.07
8/13-19	SS	0.27±0.05	44.05± 1.94		0.06±0.04	for August	Bkgd.	0.02±0.02
	DS	0.15±0.14	1,651 ± 27.9		4.9 ±0.15		0.56±0.04	0.8±0.06
8/20-26	SS	0.07±0.04	64.3 ± 3.0		0.31±0.26		0.02±0.02	Bkgd.
	DS	0.13±0.11	1,275 ± 84		2.48±0.29		0.63±0.04	0.76±0.06
8/27-9/2	SS	0.08±0.06	18.8 ± 1.3		0.13±0.14		0.01±0.02	0.12±0.01
	DS	0.28±0.05	1,937 ± 35		2.89±0.14		1.1 ±0.07	0.37±0.04

*Bkgd. indicates background.
**Value is estimated.

Blank spaces indicate data not available.
SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
9/3-9	SS	0.14±0.04	17.8 ± 1.1		Bkgd.*		0.02±0.07	Bkgd.
	DS	0.38±0.05	1,738 ±100		1.51±0.06		0.57±0.05	0.5 ±0.05
9/10-16	SS	0.05±0.04	19.0 ± 1.2		0.22±0.01	0.04±0.02	Bkgd.	0.06±0.02
	DS	0.41±0.06	1,129 ± 89.1		2.22±0.22	0.07±0.05	1.07±0.08	0.9 ±0.06
9/17-23	SS	0.09±0.05	28.8 ± 1.58		0.17±0.15	for September	0.55±0.05	0.1 ±0.04
	DS	0.12±0.03	821.7 ± 6.93		2.0 ±0.3		0.07±0.02	0.52±0.04
9/24-30	SS	0.05±0.02	26.1 ± 1.24		0.35±0.15		0.04±0.02	0.23±0.03
	DS	0.36±0.06	1,435.5 ±147.5		1.6 ±0.17		0.32±0.03	0.05±0.05
10/1-7	SS	0.07±0.02	16.5 ± 1.2		0.23±0.03		0.08±0.02	Bkgd.
	DS	0.05±0.02	1,069.2 ±118.8		2.19±0.28		0.5 ±0.04	0.27±0.03
10/8-14	SS	0.05±0.04	10.9 ± 1.0		0.83±0.33	0.19±0.05	0.11±0.03	0.04±0.01
	DS	0.46±0.05	1,590 ±115		4.85±0.22	0.13±0.04	0.39±0.11	0.36±0.04
10/15-21	SS	0.21±0.04	17.2 ± 1.4		0.30±0.15	for October	0.31±0.02	0.03±0.02
	DS	0.46±0.70	1,810 ±195		2.41±0.32		0.5**	1.35±0.11
10/22-28	SS	0.14±0.09	9.3 ± 0.9		Bkgd.		0.15±0.06	0.05±0.02
	DS	0.4 ±0.04	1,530 ±116		1.41±0.17		1.03±0.07	0.98±0.12
10/29-11/4	SS	0.2 ±0.02	27.1 ± 1.46		0.33±0.23		0.03±0.02	Bkgd.
	DS	0.45±0.08	1,439 ±113		2.16±0.08		3.9 ±0.2	1.24±0.1

*Bkgd. indicates background.

**Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
11/5-11	SS	0.14±0.02	6.0 ± 2.34		2.0 ±0.03		TS 0.33±0.0	TS 0.94±0.12
	DS	0.4 ±0.06	1,538 ±197		6.0 ±0.5			
11/12-18	SS	0.1 ±0.03	11.4 ± 1.04		1.46±0.12	0.02±0.02	0.3 ±0.2	TS 0.7 ±0.1
	DS	0.56±0.05	1,640 ±235		6.0 ±0.44	0.33±0.07	0.4 ±0.05	
11/19-25	SS	0.09±0.04	253 ± 6.0		1.1*	for November	TS 0.5*	TS 0.9 ±0.01
	DS	0.6 ±0.02	1,560 ±145		1.9 ±0.3			
11/26-12/2	SS	Bkgd.**	20.4 ± 2.5		0.5 ±0.17		0.03±0.01	0.2 ±0.01
	DS	0.4 ±0.02	1,366 ± 89.1		2.6 ±0.09		0.4 ±0.07	0.7 ±0.06
12/3-9	SS	0.07±0.01	8.0 ± 0.73		0.2 ±0.04		0.06±0.04	0.04±0.02
	DS	0.4 ±0.07	1,564 ±102		2.8 ±0.1		0.54±0.07	1.0 ±0.01
12/10-16	SS	0.2 ±0.03	7.2 ± 0.9		0.87±0.03	0.06±0.03	0.03±0.02	0.07±0.02
	DS	0.35±0.01	782 ± 26.3		1.3 ±0.4	0.6 ±0.2	1.9 ±0.2	0.8 ±0.07
12/17-23	SS	0.16±0.04	3.89± 0.57		0.22±0.05	for December	0.08±0.02	0.04±0.03
	DS	0.4 ±0.1	454 ± 19.2		4.77±0.7		0.8 ±0.1	0.82±0.14
12/24-30	SS	0.06±0.03	Bkgd.		0.07±0.03		0.05±0.02	0.05±0.03
	DS	0.12±0.07	879 ± 19		2.29±0.32		0.67±0.05	0.06±0.02

*Value is estimated.

**Bkgd. indicates background.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

CRM 41.5 WOC mile 0.6 CRM 14.6 CRM 5.5 TR529.9

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
12/31-1/6	SS	0.05±0.03	6.4 ± 1.2		0.03±0.02		0.07±0.02	Bkgd.*
	DS	0.5 ±0.08	816 ± 28		1.7 ±0.36		1.4 ±0.17	0.81±0.18
1/7-13	SS	0.07±0.06	5.21± 0.7	0.2 ±0.04	0.4 ±0.03	0.13±0.04	0.08±0.03	0.35±0.02
	DS	0.7 ±0.07	636 ± 14.3	1.6 ±0.3	0.79±0.07	0.5 ±0.02	0.71±0.10	0.7 ±0.1
1/14-20	SS	0.07±0.03	5.3 ± 2.4	0.9 ±0.07	0.06±0.06	for January	Bkgd.	0.04±0.02
	DS	0.4 ±0.01	719 ± 28.2	6.20±0.8	1.52±0.07		0.95±0.14	0.70±0.12
1/21-27	SS	0.1 ±0.03	10.28± 0.96	0.22±0.04	0.03±0.01		0.29±0.03	Bkgd.
	DS	0.53±0.13	714 ± 35.3	5.0 ±0.8	4.2 ±0.4		0.57±0.13	0.59±0.10
1/28-2/3	SS	0.1 ±0.04	6.87± 0.83	0.49±0.17	TS 1.6**		0.05±0.01	0.1 ±0.03
	DS	0.53±0.04	572 ± 17	1.01±0.54			0.6 ±0.1	1.34±0.22
2/4-10	SS	0.12±0.03	13.87± 1.54	0.02±0.02	TS 1.3**	0.46±0.23	0.06±0.02	0.03±0.02
	DS	0.4 ±0.01	1,129 ± 42	1.0 ±0.2		0.7 ±0.5	0.92±0.43	0.14±0.08
2/11-17	SS	0.03±0.02	6.6 ± 1.07	0.12±0.03	0.21±0.01	for February	0.03±0.02	Bkgd.
	DS	0.2 ±0.04	1,285 ± 56.6	1.54±0.71	1.21±0.34		0.8 ±0.3	0.6 ±0.2
2/18-24	SS	0.28±0.04	20.1 ± 1.2	0.13±0.03	0.09±0.02		0.11±0.02	0.03±0.02
	DS	0.8 ±0.02	577 ± 7.28	3.7 ±0.4	2.0 ±0.53		3.0 ±0.3	2.18±0.43
2/25-3/3	SS	0.22±0.05	8.5 ± 0.94	0.15±0.11	0.14±0.06		0.05±0.02	0.25±0.03
	DS	0.28±0.27	122.9 ± 5.35	2.5 ±0.3	7.5 ±0.2		1.48±0.21	5.4 ±0.4

*Bkgd. indicates background.

**Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
3/4-10	SS	0.1 ±0.03	8.8 ± 0.9	0.08±0.03	0.2 ±0.04		Bkgd.*	0.04±0.02
	DS	0.9 ±0.2	636 ± 33.3	1.9 ±0.04	1.4 ±0.2		0.73±0.04	1.18±0.2
3/11-17	SS	0.84±0.03	TS 1,500**	0.16±0.06	0.1 ±0.05	0.05±0.02	0.05±0.03	0.03±0.01
	DS	0.6 ±0.01		1.1 ±0.1	0.85±0.12	0.37±0.04	0.23±0.06	0.3 ±0.04
3/18-24	SS	0.16±0.03	18.4 ± 1.22	0.09±0.04	0.15±0.03	for March	0.03±0.02	0.03**
	DS	0.6 ±0.07	1,101 ± 97.8	2.2 ±0.2	2.3 ±0.39		1.8 ±0.2	1.7 ±0.3
3/25-31	SS	0.04±0.02	4.28± 0.81	0.19±0.05	0.09±0.05		0.06±0.02	0.04±0.02
	DS	0.8 ±0.15	1,420 ±101	4.8 ±0.2	3.1 ±0.2		1.09±0.1	1.19±0.08
4/1-7	SS	0.08±0.03	5.08± 1.01	0.2**	0.09±0.02		0.05±0.02	0.09±0.02
	DS	0.6 ±0.1	1,600**	10.0 ±0.55	0.62±0.52		1.4 ±0.2	1.51±0.21
4/8-14	SS	0.2 ±0.05	6.85± 0.75	0.05±0.05	Bkgd.	0.05±0.04	0.05±0.02	0.02±0.01
	DS	1.05±0.25	527 ± 12	6.0 ±0.6	13.32±1.81	0.62±0.17	3.36±0.3	1.21±0.15
4/15-21	SS	0.04±0.02	12.1 ± 1.7	Bkgd.	0.12±0.05	for April	0.17±0.03	0.02**
	DS	0.4 ±0.04	1,368 ± 14	8.45±0.28	7.49±0.22		1.61±0.16	2.04±0.15
4/22-28	SS	0.05±0.02	10.4 ± 0.91	0.07±0.02	Bkgd.		0.04±0.02	0.03±0.01
	DS	1.57±0.10	2,000**	4.45±0.20	2.94±0.42		1.85±0.12	4.45±0.2
4/29-5/5	SS	0.04±0.01	18.75± 1.53	0.06±0.02	0.16±0.03		0.05±0.03	0.04±0.03
	DS	0.87±0.08	2,700**	11.61±0.46	1.99±0.42		4.15±0.14	1.11±0.07

*Bkgd. indicates background.

**Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
5/6-12	SS	0.05±0.02	15.0 ± 1.06	0.83±0.03	0.11±0.02		0.04*	0.05*
	DS	0.51±0.09	2,852 ±178	2.17±0.37	1.85±0.36		1.34±0.07	1.07±0.14
5/13-19	SS	0.06±0.02	13.8 ± 1.4	0.09±0.02	0.09±0.02	0.05±0.02	Bkgd.**	0.06±0.02
	DS	0.54±0.11	2,700*	3.75±0.43	4.85±0.43	0.99±0.17	2.35±0.31	1.02±0.13
5/20-26	SS	0.02±0.02	25.2 ± 1.44	0.11±0.03	0.11±0.07	for May	0.05±0.02	0.29*
	DS	0.72±0.19	2,400 ±144	2.55±0.35	4.23±0.28		0.08±0.01	0.99±0.13
5/27-6/2	SS	0.06±0.02	21.8 ± 1.29	0.16±0.03	0.12±0.03		0.02±0.02	0.17*
	DS	0.62±0.16	1,900*	4.27±0.53	3.3 ±0.81		1.11±0.14	1.58±0.16
6/3-9	SS	0.07±0.03	2.85± 0.15	0.33±0.01	0.2*		0.05±0.02	0.06±0.02
	DS	0.87±0.13	1,579 ±122.5	8.5 ±0.65	4.9 ±0.53		1.23±0.16	1.26±0.25
6/10-16	SS	0.25±0.10	28.1 ± 3.23	0.22±0.04	0.21±0.07	0.04±0.02	Bkgd.	Bkgd.
	DS	0.4 ±0.08	1,258 ± 92.3	3.7 ±0.28	2.8 ±0.2	1.21±0.17	1.8 ±0.18	1.55±0.20
6/17-23	SS	0.15±0.03	23.5 ± 1.23	0.14±0.03	0.68*	for June	0.02±0.01	0.04±0.02
	DS	0.6 ±0.1	2,135 ±155.7	2.6 ±0.2	2.52±0.56		4.7 ±0.68	0.96±0.41
6/24-30	SS	Bkgd.	92.5 ± 2.48	0.24±0.04	0.99±0.07		0.11±0.03	0.05±0.03
	DS	1.2 ±0.1	1,700 ±112	2.43±0.38	3.17±0.44		1.4 ±0.2	1.5 ±0.3

*Value is estimated.

**Bkgd. indicates background.

SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
7/1-7	SS	0.10±0.02	126 ± 3.11	0.27±0.07			0.04±0.02	0.06±0.02
	DS	0.6 ±0.09	1,450 ±128	3.2*			1.85±0.22	1.0 ±0.1
7/8-14	SS	0.12±0.03	13.4 ± 1.1	0.19±0.03		0.05±0.02	0.15±0.03	0.06±0.03
	DS	0.58±0.10	865 ± 70.4	7.05±0.61		1.18±0.16 for July	1.4 ±0.16	1.1 ±0.1
7/15-21	SS	0.23±0.06	15.38± 1.03	0.05±0.02	0.03±0.02		0.07±0.06	0.5 ±0.03
	DS	0.56±0.11	1,532 ± 83	2.02±0.14	1.62±0.13 for July 1-21		1.12±0.11	1.02±0.16
7/22-28	SS		27.80± 1.58					
	DS		1,004 ± 58.7					
7/29-8/4	SS		16.55± 1.43					
	DS		1,972 ±219					
8/5-11	SS		3.63± 0.65					
	DS		1,008 ± 80.2					
8/12-18	SS	0.05±0.02	13.62± 1.10	0.02±0.01	0.11*	Bkgd.**	0.07±0.04	0.12±0.05
	DS	0.65±0.12 for July 22-August 18	1,115 ± 81.8	2.16±0.14 for July 22-August 18	1.37±0.11 for July 22-August 18	0.72±0.13 for August	2.57±0.25	1.0 ±0.12
8/19-25	SS		16.81± 0.92					
	DS		1,985 ±124					
8/26-9/1	SS		10.90± 0.83					
	DS		1,400 ±183					

*Value is estimated.

**Bkgd. indicates background.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
9/2-8	SS		16.95 ± 1.30					
	DS		985 ± 79					
9/9-15	SS	0.01±0.01	14 ± 0.64	0.03±0.01	0.01±0.01	0.02±0.02	0.10±0.02	0.10±0.03
	DS	0.75±0.09	1,002 ±193	2.14±0.14	1.94±0.14	0.55±0.08	1.45±0.15	1.15±0.11
9/16-22	SS	for Aug. 19-Sept. 15	22.28 ± 1.32	for August 19-Sept. 15	for August 19-Sept. 15	for September		
	DS		1,133 ±168					
9/23-29	SS		16.10 ± 1.10					
	DS		1,573 ±214					
9/30-10/6	SS		14.60 ± 0.89					
	DS		997 ±198					
10/7-13	SS	0.20±0.06	9.38 ± 0.76	0.03±0.01	0.01±0.01		0.03±0.02	0.53±0.05
	DS	0.76±0.09	1,226 ± 97.2	1.93±0.14	2.29±0.26		0.88±0.09	1.13±0.12
10/14-20	SS	for Sept. 16-Oct. 13	2.66 ± 0.79	for Sept. 16-Oct. 13	for Sept. 16-Oct. 13	0.04±0.02		
	DS		986 ±161			0.38±0.08		
10/21-27	SS		0.50 ± 0.38			for October		
	DS		1,171.2 ± 72.4					

Blank spaces indicate data not available.
SS = suspended solids; DS = dissolved solids.

Table 1 (Continued)

CONCENTRATIONS OF STRONTIUM-90, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
10/28-11/3	SS		2.86 ± 0.45					
	DS		1,326 ±178					
11/4-10	SS	0.25±0.05	1.79 ± 0.42	0.03±0.01	0.04±0.01		0.03±0.02	0.14±0.03
	DS	0.94±0.13	500 ±124	4.36±0.28	2.36±0.25		0.01±0.01	1.11±0.12
		for Oct. 14-Nov. 10		for Oct. 14-Nov. 10	for Oct. 14-Nov. 10			
11/11-17	SS		1.46 ± 0.38			0.08±0.03		
	DS		848 ± 73.2			0.81±0.12		
						for November		
11/18-24	SS		7.69 ± 0.75					
	DS		236 ± 26.3					
11/25-12/1	SS		8.00 ± 1.60					
	DS		1,155 ±164.0					

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

To determine what portion of the total strontium-90 activity is associated, on the average, with the suspended solids, and what portion with the dissolved solids (meaning in solution and/or associated with very fine suspended particles not removed by the supercentrifuge), a simple average percentage was computed for each of the two portions from the determinations on all samples from each station, with results as shown in the following tabulation. Median values are also shown.

Distribution of Strontium-90 in Water Samples

Station	Percent Total Activity in			
	Suspended Solids		Dissolved Solids	
	Mean	Median	Mean	Median
Clinch River at Oak Ridge water plant	24	21	76	79
White Oak Creek at White Oak Dam	2	1	98	99
Clinch River at Gallaher Bridge	6	4	94	96
Clinch River at Centers Ferry	9	6	91	94
Tennessee River at Watts Bar Dam	9	6	91	94
Tennessee River at Chickamauga Dam	10	6	90	94

From these data it is quite apparent that from 90 to 98 percent of the strontium-90 activity is associated with the dissolved solids, or in other words, dissolved in the water itself. (The maximum size of sediment particles left in suspension by the supercentrifuge is estimated to be 0.7 microns.) The time of contact with the suspended solids in Clinch River appears to have some influence on the distribution of activity between suspended solids and dissolved solids since the percentage associated with the dissolved solids decreases from 98 percent at White Oak Dam to 94 percent at Gallaher Bridge, and to 91 percent at Centers Ferry. There is essentially no change, however, from Centers Ferry to Chickamauga Dam.

Mass Curves--Mass (cumulative) curves of strontium-90 loads at all stations except Loudon are shown in figure 4. The Loudon loads have been computed on the assumption that the concentrations found represent flow-weighted concentrations. These monthly loads are shown in figure 4 by the vertical bars extending up from the Centers Ferry load. In preparing all the mass curves in this report, the total activity in each sample was used in the computations; i.e., the total sample activity is determined as the sum of the activities in both the suspended and dissolved solids.

To permit comparison of the total cumulative loads, at successive stations, an estimate of the "normal" time of water travel from station to station was made and lagged time scales were used for plotting the loads accordingly. For example, water flowing out of the mouth of White Oak Creek would be expected to arrive at the Centers Ferry station

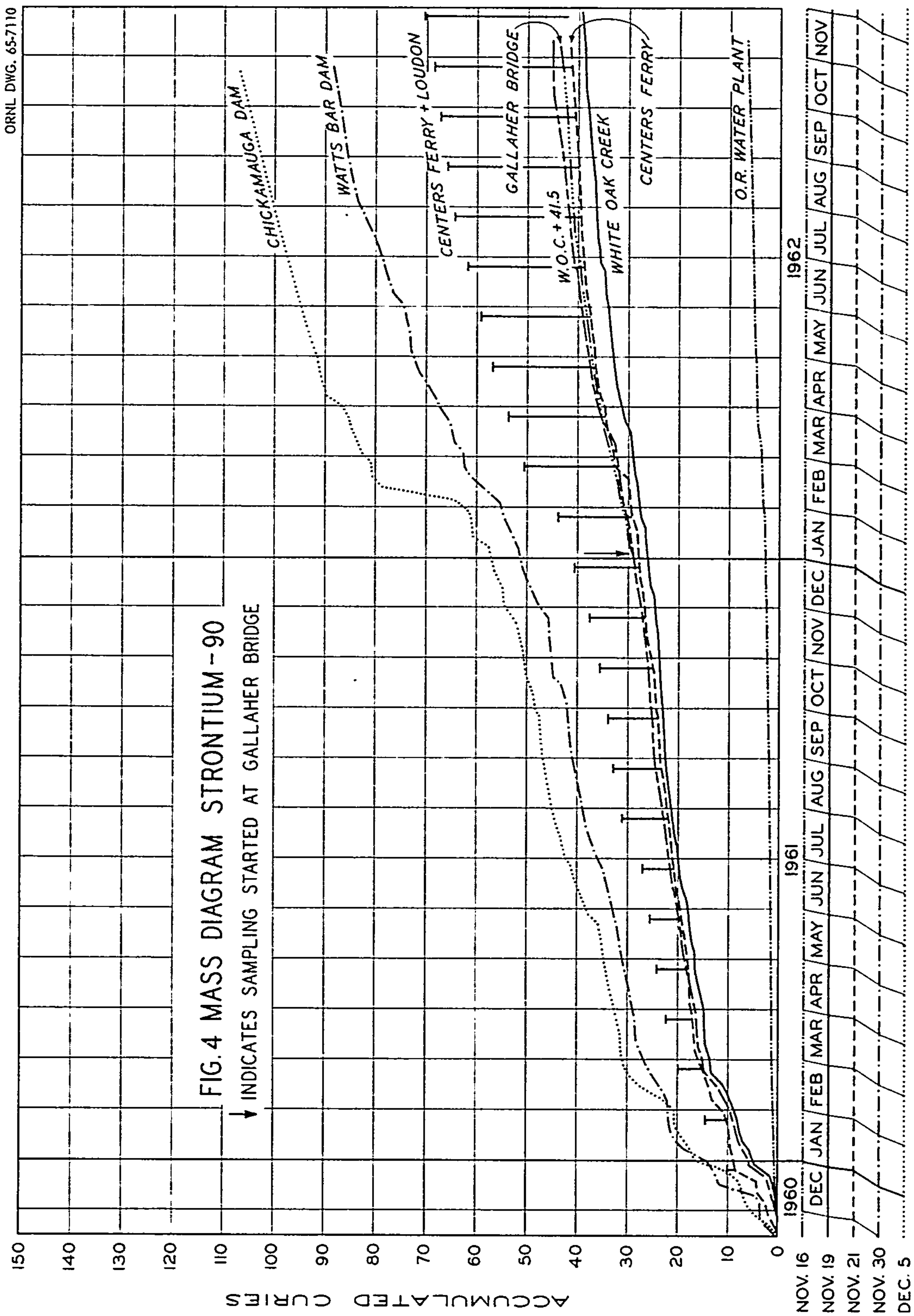

FIG. 4 MASS DIAGRAM STRONTIUM-90

(Clinch River mile 5.5) two days later, and to arrive at Watts Bar Dam nine days later, and at Chickamauga Dam after five more days. Naturally, these times are not constant but vary with streamflows, pool levels, and to some extent with the season of the year. A constant time of travel has been assumed, however, as detailed above, and the plotted data seem to support, over all, the estimated times reasonably well.

The rate of discharge of strontium-90 to Clinch River during the first three months of the sampling period was approximately 4.5 curies per month but at the end of February 1961 the rate was abruptly reduced to approximately 1.2 curies per month, a rate that was maintained quite uniformly throughout the remainder of the two-year sampling period.

The strontium-90 load measured for the Clinch River at the Oak Ridge water plant is also shown in figure 4. The rate of accumulation was quite steady throughout the two-year period, at about 0.29 curies per month, or at about 91 microcuries per square mile per month.

Combining the sum of the two loads, White Oak Creek and Clinch River at Oak Ridge water plant, produces a third curve, also shown in the figure. The curve representing the sum of these two loads exceeds, at the end of sampling, the load measured at Centers Ferry by approximately 13 percent. There is also a slight loss indicated between the stations at Gallaher Bridge and Centers Ferry during the 11 months of record at Gallaher Bridge. Since about 9 percent of the total strontium-90 activity at the Centers Ferry station is associated with the suspended solids, the apparent explanation for part of this loss is sedimentation in the embayment of Clinch River.

Although the daily Loudon samples were not proportioned to streamflow, if it is assumed that they were, a sizable load presumably derived from fallout is shown as flowing down the Tennessee River from the 12,220 square miles of drainage area above this station. The accumulated load, with the above assumption, was found to be about 28 curies representing a contribution, averaged over the 24 months of record, of 96 microcuries per square mile per month.

Additional gains are indicated from the 1,550 square mile drainage area between Centers Ferry plus Loudon and Watts Bar Dam. This gain was 21 curies representing a contribution of approximately 590 microcuries per square mile per month. On the assumption that the Loudon load is not correct (and since the samples were not flow-proportioned, it undoubtedly is not), if the Centers Ferry load is subtracted from the Watts Bar load the contribution per square mile from the intervening area is determined to be 151 microcuries per square mile per month.

There is a large increase in the load at Chickamauga Dam during late February 1962, presumably from fallout following numerous bomb tests (Russian and some American) during the months of September, October, and November 1961. The total accumulated increase between Watts Bar and Chickamauga Dams in the two-year period was approximately 19 curies, representing a contribution of 227 microcuries per square mile per month.

Downstream from the Centers Ferry station, there was a measured gain in the total load from station to station. However, this cannot be interpreted to mean that all the strontium-90 originating at Oak Ridge is transported past Chattanooga, Tennessee. All that can be said on this point, with a reasonable degree of confidence, is that a large percentage of the Oak Ridge load does pass Chattanooga. That quantity of strontium-90 lost from solution and suspension during the two-year sampling period was apparently more than offset by contributions to the river system from fallout. In fact, the two-year load at Chattanooga is over two and one-half times the load passing White Oak Dam.

Periods of 1961 and 1962 in which fairly frequent nuclear bomb detonations occurred in both the United States and in the USSR, together with the resultant effects on gross beta concentrations at eight remote (remote from Oak Ridge) precipitation stations are shown in figure 5. (These data were supplied by the Applied Health Physics Section.) The stations were located at Norris, Fort Loudon, Douglas, Cherokee, Watts Bar, Great Falls, and Dale Hollow Dams, and at Berea, Kentucky.

The abrupt increases in the strontium-90 loads during February 1962 at Loudon, Watts Bar Dam, and at Chickamauga Dam would appear to reflect relatively large volumes of runoff containing strontium-90.

Comparison with Load Measured by ORNL--To determine how strontium-90 loads at White Oak Dam, as measured in this study, compare with the same loads measured by the Oak Ridge National Laboratory, these two sets of data were plotted by months in figure 6. Reported loads for several of the individual months are greatly different but for the two years of record, the total load as determined by ORNL was only about 12 percent less than that measured in the Clinch River Study.

Cesium-137, Concentrations and Total Stream Loads

Concentrations of cesium-137 found in all samples at all stations for the two-year period of record are shown in table 2. However, because of extreme difficulty in analyzing the gamma spectrum to identify the activity due strictly to cesium-137 when there is a high concentration of ruthenium-106 present, the data reported here on cesium-137 must be considered only very approximate. Cumulative loads may be reasonably correct (due to tendency for positive and negative errors to balance out), but no great confidence can be placed in any of the cesium-137 data. In retrospect it can be said that the cesium should have been separated chemically or by other means from the samples before any radiological determinations were made.

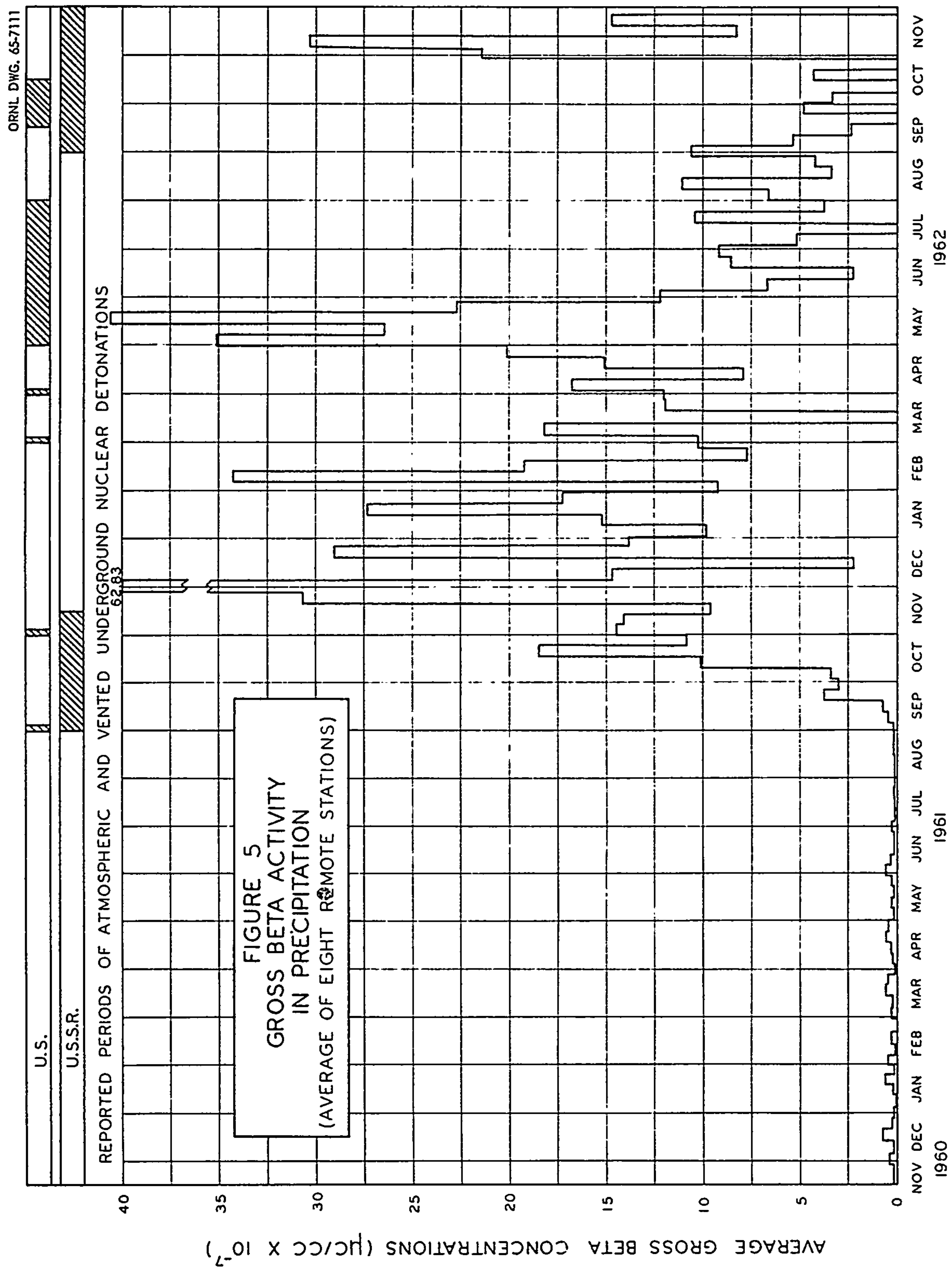

FIGURE 5
GROSS BETA ACTIVITY
IN PRECIPITATION
(AVERAGE OF EIGHT REMOTE STATIONS)

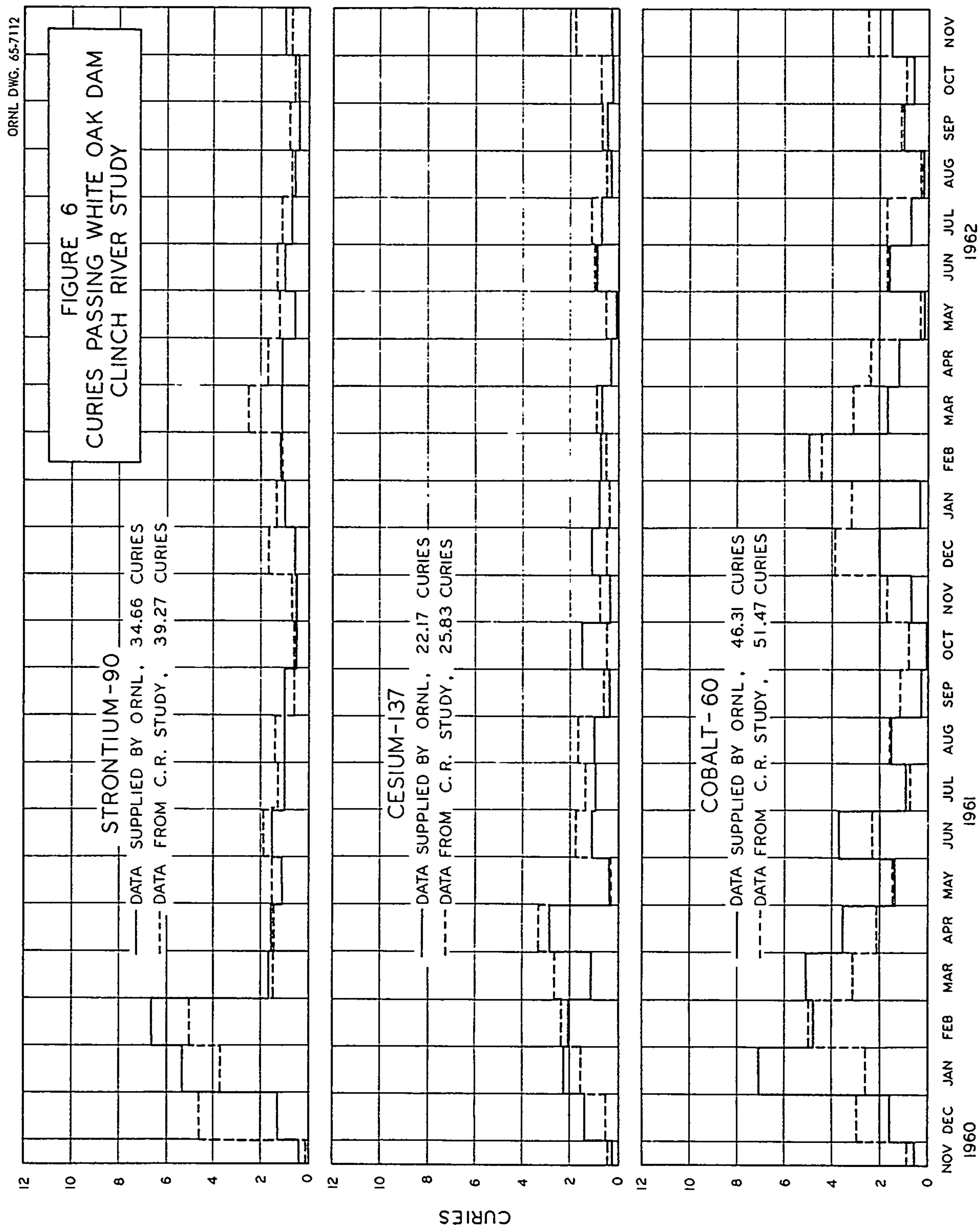

FIGURE 6
CURIES PASSING WHITE OAK DAM
CLINCH RIVER STUDY

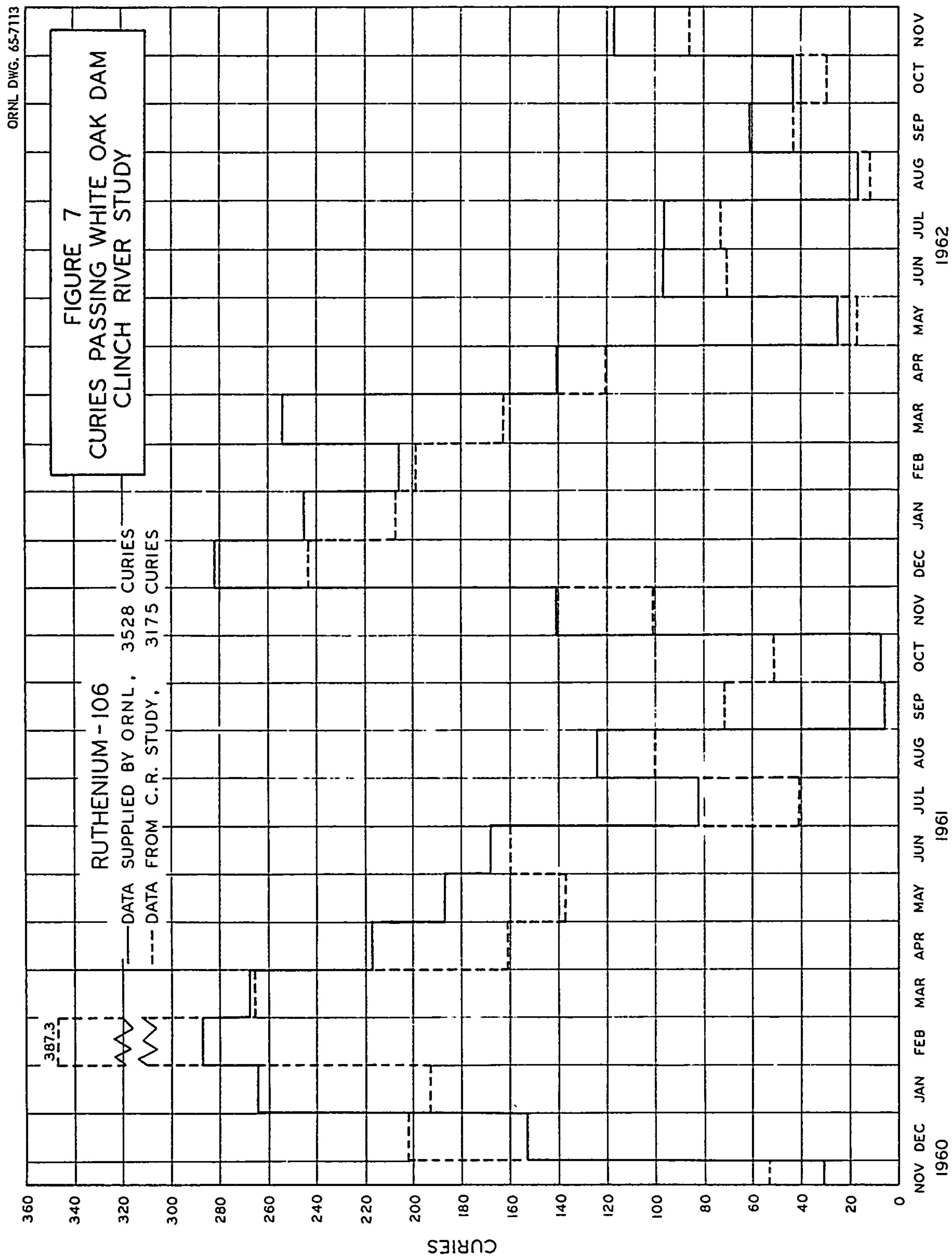

FIGURE 7
CURIES PASSING WHITE OAK DAM
CLINCH RIVER STUDY

Table 2

CONCENTRATIONS OF CESIUM-137, pc per liter

Date		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
1960								
11/13-19	TS	1	978		6			
11/20-26	TS	1	-36		11			1
11/27-12/3	TS	1	4,225		6*		2	1
12/4-10	TS	2	778		4		1	0
12/11-17	TS	0	190		-7	10	1	6
12/18-24	TS	1	316		4	for December	1	2
12/25-31	TS	0	74		22		0	2
1961								
1/1-7	TS	1	696		6		0	0
1/8-14	TS	0	688		1	1	-1	0
1/15-21	TS	2	1,383		7	for January	0	0
1/22-28	TS	0	180		3		0	0
1/29-2/4	TS	3	666		1		-2	0
2/5-11	TS	1	2,978		-4		0*	0
2/12-18	TS	2	824		8	34	1	1
2/19-25	TS	5	787		3*	for February	0	0
2/26-3/4	TS	3*	366		4		2	2
3/5-11	TS	2	523		6		2	0
3/12-18	TS	1	2,082		10	SS 1	1	3
3/19-25	TS	5	2,742		6	DS 1	4	2
3/26-4/1	TS	5	1,500*		17	for March	2	4
4/2-8	TS	2	329		4		7	1
4/9-15	TS	0	4,292		1		0	0
4/16-22	TS	0	462		0	SS 0	0	0
4/23-29	SS	0*	832		5	DS 3	TS 0	TS 0
	DS	0	740		0*	for April		

*Value is estimated.
Blank spaces indicate data not available. TS = total solids; SS = suspended solids; DS = dissolved solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
4/30-5/6	SS	0	667		3		0	1
	DS	0	43		0*		0	1
5/7-13	SS	0	377		2	0	0	0
	DS	0	0*		0*	0 for May	1	0
5/14-20	SS	0*	184		1		0	0
	DS	0	0		0		0	0
5/21-27	SS	0	341		5		0	0
	DS	0	38		1		0	0*
5/28-6/3	SS	0	632		6		0	0
	DS	0	0		0		0	0
6/4-10	SS	0	1,180*		6	0	0	0
	DS	0	0		1	0 for June	0	0
6/11-17	SS	0	1,052		7		0	0
	DS	0	-70		0		0	1
6/18-24	SS	1	747		16		0	0
	DS	0	1,310*		-3		0	0
6/25-7/1	SS	1	4,365		13		0	0
	DS	0	426		0		0	0

*Value is estimated.

Blank spaces indicate no data available.

SS = suspended solids; DS = dissolved solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
7/2-8	SS	6	703		1		0	0
	DS	0	87		0		0	0
7/9-15	SS	0	966		3	0	0	0
	DS	0	5,443		0	0	0	0
7/16-22	SS	0	731		4	for July	0	0
	DS	0	1,019*		0		0	0
7/23-29	SS	0	1,190		6		0	0
	DS	0	-65		1		0	0
7/30-8/5	SS	0	1,585		2		0	0
	DS	0	1,178		1		0	0
8/6-12	SS	0	2,188		8	1	0	0
	DS	0	1,492		5	6	0	0
8/13-19	SS	3	1,445		0	for August	0	0
	DS	3	-253		0		0	0
8/20-26	SS	0	1,061		4		0	0
	DS	0	-221		0		0	0
8/27-9/2	SS	0	484		1		0	0
	DS	2	1,341*		0		0	3

*Value is estimated.

Blank spaces indicate no data available.

SS = suspended solids; DS = dissolved solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
9/3-9	SS	0	TS 1,880*		1		0	2*
	DS	0			0		0	0
9/10-16	SS	0	TS 1,290*		2	0	0	0
	DS	0			0	-2	0	0
9/17-23	SS	0	TS 1,320*		2	for September	0	0
	DS	0			0		0	0
9/24-30	SS	0	TS 1,145*		19		0	0
	DS	0			1		0	0*
10/1-7	SS	0	636		12		0	0
	DS	0	-265		2		0	0
10/8-14	SS	0	571		49	0	0	0
	DS	2	25		6	0	0	1
10/15-21	SS	0	985		29	for October	0	0
	DS	0	315*		0		0	1
10/22-28	SS	0	1,860*		18		0	0
	DS	0	131		1		1	0
10/29-11/4	SS	0	1,156		30		0	0
	DS	0	885		4		0	0

*Value is estimated.

Blank spaces indicate no data available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
11/5-11	SS	0	106		116		TS 0	TS 0
	DS	0	1,578		8			
11/12-18	SS	0	823		94	0	0	TS 0
	DS	0	2,330		2	0*	0	
11/19-25	SS	0	159		75*	for November	TS 0*	TS 0
	DS	-2	553		1			
11/26-12/2	SS	0	349		39		0	0
	DS	0	223		1		0	0
12/3-9	SS	0	379		3		0	0
	DS	0	595		2		0	0
12/10-16	SS	0	321		5	0	0	0
	DS	0	0*		-6	0	-2	0
12/17-23	SS	0	96		5	for December	0	1
	DS	-1	0*		0*		-3	-1
12/24-30	SS	0	70		2		0	0
	DS	-1	380*		-6		18	-3

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
12/31-1/6	SS	0	88		1		0	0
	DS	-4	275*		-8		-2	-1
1/7-13	SS	0	71	1	1	0	0	0
	DS	-4	320*	-2	1	-1	-5	-3
1/14-20	SS	0	153	2	2	for January	0	0
	DS	-4	194	-2	-6		0*	0*
1/21-27	SS	0	288	6	2		0	0
	DS	0*	0*	-1	0*		-2	-3
1/28-2/3	SS	0	286	8	TS 1*		0	0
	DS	0*	0*	1			-3	1
2/4-10	SS	-1	308	4	TS 1*		0	0
	DS	0*	170*	-1			0*	-1
2/11-17	SS	0	153	5	3	0	0	0
	DS	-1	430*	-1	-2	-1	0*	0*
2/18-24	SS	-1	590	7	2	for February	0	0
	DS	-9	0*	0	3*		-2	1
2/25-3/3	SS	0	350	5	4		0	0
	DS	0	0*	-2	-2		-4	1*

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
3/4-10	SS	2	301	4	7		0	1
	DS	0	120*	0	1		3*	-4
3/11-17	SS	1	TS 630*	4	6	0	0	1*
	DS	0		-1	0	-1	5	0
3/18-24	SS	1	450	-6	5	for March	3*	0
	DS	-2	75	-4	-1		-1	-1
3/25-31	SS	0	98	5	12		1	0
	DS	-2	197	-3	-3		-1	0
4/1-7	SS	0	141	0	7		0	0
	DS	0	59	4	-2		-2	0
4/8-14	SS	2	203	TS 3*	5	1	0	0*
	DS	-1	0*		1	-1	-2	-3
4/15-21	SS	1	323	5	5	for April	1	0
	DS	0	15*	1	0		0	-2
4/22-28	SS	1	388	2	4		1	0
	DS	0	200*	-1	0		1	-1
4/29-5/5	SS	0	530*	1	2		0	0
	DS	0	288	0	0		0	1

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
5/6-12	SS	1	742	2	2		0	0
	DS	-1	202	0	0		0	-2
5/13-19	SS	0	595	4	3	0	0	0
	DS	-1	395*	-1	0	0	0	0
						for May		
5/20-26	SS	0	973	5	5		0	0
	DS	0*	225*	-2	-2		-1	0
5/27-6/2	SS	0*	1,283	8	7		0	0
	DS	0*	1	0	-1		-1	-1
6/3-9	SS	0	977	9	11		1	0
	DS	-1	425*	0	5		0	-1
6/10-16	SS	0	927	12	5	0	0	0
	DS	-3	250	1	1	-1	-1	-2
						for June		
6/17-23	SS	0	929	9	22*		0	0
	DS	-1	1,370*	0	-1		0	-1
6/24-30	SS	1	1,070	16	34		0	0
	DS	0	0*	0	1		-1	-1
7/1-7	SS	1	816	15	TS 16*		0	0
	DS	-1	410*	0*			0	-1

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
7/8-14	SS	1	613	18	TS 10*		0	0
	DS	0	370	-1			0	0
7/15-21	SS	0	1,194	8	0	0	0	0
	DS	0	38	1	4	1	-1	0
7/22-28	SS	0*	1,129	3	4*	for July	0*	0
	DS	0	538	0	0		0	-1
7/29-8/4	SS	1	955	1	TS 3*		0	0
	DS	0	68	0			0	0
8/5-11	SS	2	781	TS 24*	TS 49*		0	0
	DS	0	295*				0	1
8/12-18	SS	1	1,124	4	5	0	0	0
	DS	2	155*	0	0	0	0	-3
8/19-25	SS	1	1,210	3	3	for August	0	0*
	DS	1	27	0	1		1	1
8/26-9/1	SS	1	904	3	3		0	0*
	DS	-1	59	0	0		1	0*
9/2-8	SS	0	966	2	5		0	0
	DS	0	73	0	1		1	0

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
9/9-15	SS	1	720	6	3		0	0
	DS	0	240*	-5	1		0	0
9/16-22	SS	1	610	8	1	0	0	0
	DS	0	0*	-9	8	0	1	0
9/23-29	SS	0	766	4	3	for September	0	0
	DS	0	970*	0	5		0	0
9/30-10/6	SS	0	710	4	4		2	0
	DS	0	570*	2	0		1	0
10/7-13	SS	1	1,000	4	4		0	0
	DS	0	730	0	4		1	1
10/14-20	SS	0	493	9	5	0	0	0
	DS	0	115	2	0	1	0	-1
10/21-27	SS	0*	280	12	2	for October	0	0
	DS	1	695*	0	1		3	0
10/28-11/3	SS	0	308	11	2		0	0
	DS	1	1,820*	2	1		0	0

*Value is estimated.

SS = suspended solids; DS = dissolved solids.

Table 2 (Continued)

CONCENTRATIONS OF CESIUM-137, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
11/4-10	SS	0	282	11	1		0	0
	DS	-1	884	6	1		2	3
11/11-17	SS	1	88	13	3	0	0	2*
	DS	0	2,660*	1	4	0 for November	1*	0
11/18-24	SS	0*	410	9	4		1	0
	DS	-3	656	12	0*		-1	0
11/25-12/1	SS	0	359	8	1			0
	DS	0		2	3			0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

Maximum concentrations found in the weekly (monthly at Loudon) composite samples (including both suspended and dissolved solids) are shown in the following tabulation:

Station	Sample Showing Highest Concentration Cesium-137 Concentration pc per liter	Period of Occurrence
Clinch R. at Oak Ridge water plant	6	Jul. 2-8, 1961, and Aug. 13-19, 1961
White Oak Creek at White Oak Dam	6,409	Jul. 9-15, 1961
Clinch R. at Gallaher Bridge	21	Nov. 18-24, 1962
Clinch R. at Centers Ferry	35*	Jun. 24-30, 1962
Tennessee R. at Loudon, Tenn.	34	Feb. 5-11, 1961
Tennessee R. at Watts Bar Dam	18	Dec. 24-30, 1961
Tennessee R. at Chickamauga Dam	6	Dec. 11-17, 1960

*Omitting high values during period September 10 through December 2, 1961, when sampling equipment was not functioning properly.

Since even the maximum concentrations at all stations are far below MPC values for drinking water used by the general population, mean concentrations at the various stations were not computed.

To determine what portion of the total cesium-137 activity is associated, on the average, with the suspended solids, and what portion with the dissolved solids (including, of course, those very fine suspended solids not removed by the supercentrifuge), a simple average percentage was computed for each of the two portions from the determinations made on all samples from each station, with results as shown in the following tabulation. Median concentrations are also indicated.

Distribution of Cesium-137 in Water Samples

Station	Percent Total Activity in Suspended Solids		Dissolved Solids	
	Mean	Median	Mean	Median
Clinch R. at Oak Ridge water plant	82	100	18	0
White Oak Creek at White Oak Dam	69	79	31	21
Clinch River at Gallaher Bridge	92	100	8	0
Clinch River at Centers Ferry	86	100	14	0
Tennessee River at Watts Bar Dam	30	0	70	100
Tennessee River at Chickamauga Dam	19	0	81	100

In marked contrast with strontium-90, the great bulk (69 to 92 percent) of the cesium-137 load is associated with the suspended solids in the water samples collected from White Oak Creek and from Clinch River. The Tennessee River samples, however, show 70 to 81 percent of the load to be in solution and/or associated with the very fine solids not removed by the supercentrifuge. This indicates that practically all the Clinch River sediment has settled by the time the Watts Bar Dam and Chickamauga Dam stations are reached and that only the very fine particulate matter and its contained activity remains.

Mass Curves--Mass curves of cesium-137 loads at all stations except Loudon, are shown in figure 8. In spite of a basic lack of accuracy in all cesium-137 determinations, the agreement shown in the discussion under "Comparison with Load Measured by ORNL," page 40, indicates the mass curve for White Oak Creek probably is reasonably accurate. The rate of discharge of cesium-137 to Clinch River was quite variable for the period November 1960 to April 1961, but thereafter, through November 1962, the rate of discharge was reasonably steady at about 0.8 curie per month.

The outstanding feature of all these curves that immediately catches the eye is the extremely great load shown for Centers Ferry in the fall of 1961. Due to a malfunctioning of the sampling equipment here during this period, as explained in detail in Progress Report No. 3, the reported load is undoubtedly incorrect. If the curve value for December 1, 1961, is adjusted to about 21 curies (the value obtained by extending the curve established prior to about October 1), and the load thereafter accumulated from this value, the entire mass curve for this station appears more reasonable, and is very similar to that for Gallaher Bridge.

Although there is considerable question about the accuracy of all cesium-137 determinations, still there is an indicated increase in the cesium-137 loads during 1962 between White Oak Creek and Gallaher Bridge that is quite substantial. Such an increase might be attributed to lack of accuracy were it not for the fact that when the Centers Ferry load is plotted in the lower position as discussed above, the Gallaher Bridge and Centers Ferry loads check each other amazingly well. This increase cannot logically be attributed to scouring of silt from the riverbed in the reach between White Oak Creek and Centers Ferry since the load seems to have increased more or less continuously throughout the year, and not just during the high river flows of January, February, and March 1962. Although limited accuracy in analysis of the cesium-137 samples casts serious doubts into the situation, and although a careful field investigation of this situation has previously been made by P. H. Carrigan and R. J. Pickering, still the indication of a sizable increase in the Clinch River load of this radionuclide at some point(s) below the mouth of White Oak Creek is sufficiently definite to warrant a "second look" by personnel familiar with the possibilities of seepage from disposal pits, and with all other possible sources of this radionuclide.

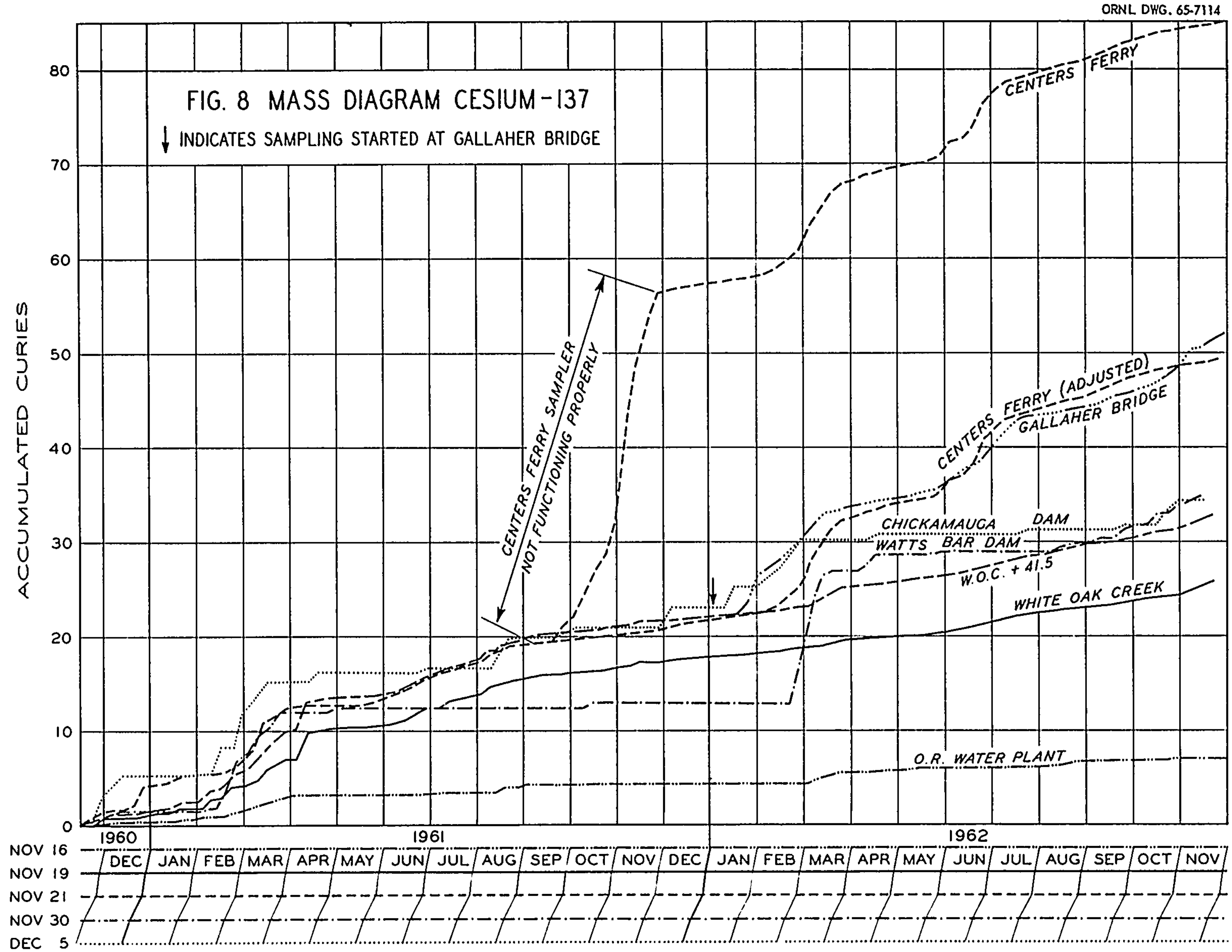

FIG. 8 MASS DIAGRAM CESIUM-137

↓ INDICATES SAMPLING STARTED AT GALLAHER BRIDGE

Because of the very limited accuracy of analysis, particularly in the dilute samples collected from the Tennessee River, no detailed discussion of the mass curves for Watts Bar and Chickamauga Dams is warranted.

Comparison with Load Measured by ORNL--The data for cesium-137 plotted in figure 6 indicate reasonable agreement during most months of record between the loads as determined by the Oak Ridge National Laboratory and as determined in the Clinch River Study. The total load for the two-year period as determined by ORNL (22.17 curies) was about 14 percent less than that determined in the Clinch River Study (25.83 curies).

Cobalt-60, Concentrations and Total Stream Loads

Concentrations of cobalt-60 found in all samples at all stations for the two-year period of record are shown in table 3.

Maximum concentrations found in the weekly (monthly at Loudon) composite samples (including activity in both suspended and dissolved solids) are shown below:

	Sample Showing Highest Concentration Cobalt-60	
Station	Concentration pc per liter	Period of Occurrence
Clinch River at Oak Ridge water plant	5	Jul. 22-28, 1962
White Oak Creek at White Oak Dam	5,095	Nov. 12-18, 1961
Clinch River at Gallaher Bridge	18	Nov. 18-24, 1962
Clinch River at Centers Ferry	52	Jun. 11-17, 1961
Tennessee River at Loudon, Tenn.	1	*
Tennessee River at Watts Bar Dam	3	**
Tennessee River at Chickamauga Dam	3	Feb. 18-24, 1962, Jun. 17-23, 1962, and Aug. 12-18, 1962

*This value occurred in several samples throughout the sampling period.

**This value occurred five times, March to October 1962, inclusive.

Table 3

CONCENTRATIONS OF COBALT-60, pc per liter

Date	Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
1960							
11/13-19	TS 0	2,302		4			
11/20-26	TS 0	4,679		8		1*	0
11/27-12/3	TS 0	3,734		8		1	0
12/4-10	TS 2	2,521		3		1	1
12/11-17	TS 0	3,156		6	0	1	0
12/18-24	TS 0	3,147		12	for December	1	0
12/25-31	TS 1	3,391		12		1	0
1961							
1/1-7	TS 0	2,022		19		1	1
1/8-14	TS 0	2,246		2		1	1
1/15-21	TS 0	3,656		3	0	2	2
1/22-28	TS 0	2,533		3	for January	1	2
1/29-2/4	TS 0	3,611		2		1	2
2/5-11	TS 0	3,817		34		1*	1
2/12-18	TS 0	2,765		28	1	0	1
2/19-25	TS 0	1,784		5*	for February	2	1
2/26-3/4	TS 0*	1,418		4		1	1
3/5-11	TS 0	1,046		2		0	0
3/12-18	TS 0	1,526		4	SS 0	0	0
3/19-25	TS 0	1,861		2	DS 0	0	0
3/26-4/1	TS 0	1,657		3	for March	0	0
4/2-8	TS 0	1,612		13		0	0
4/9-15	TS 0	1,566		4	SS 0	0	0
4/16-22	TS 0	1,521		10	DS 0	1	0
					for April		

*Value is estimated.

Blank spaces indicate data not available. TS = total solids; SS = suspended solids; DS = dissolved solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
4/23-29	SS	0*	157		1			
	DS	0	852		1*		TS 1	TS 0
4/30-5/6	SS	0	254		0		0	0
	DS	0	102		1		0	0
5/7-13	SS	0	182		0	0	0	0
	DS	0	1,578		7	0	1	0
						for May		
5/14-20	SS	0*	9		0		0	0
	DS	0	1,921		4		0	1
5/21-27	SS	0	69		2		0	0
	DS	0	1,303		8		0	0*
5/28-6/3	SS	0	147		1		0	0
	DS	0	928		2		1	0
6/4-10	SS	0	940*		1	0	0	0
	DS	0	1,893		2	0	0	0
						for June		
6/11-17	SS	0	302		3		0	0
	DS	0	2,037		49		0	1
6/18-24	SS	0	240		3		0	0
	DS	0	1,831		15		0	0
6/25-7/1	SS	0	201		2		0	0
	DS	0	1,051		6		0	0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
7/2-8	SS	0	125		1		0	0
	DS	0	791		0		0	1
7/9-15	SS	0	210		0	0	0	0
	DS	0	2,104		1	0	1	1
7/16-22	SS	0	1,191		4	for July	0	0
	DS	0	0*		4		1	1
7/23-29	SS	0	151		1		0	0
	DS	0	541		2		0	0
7/30-8/5	SS	0	1,018		0		0	0
	DS	0	1,163		0		0	0
8/6-12	SS	0	344		0		0	0
	DS	0	1,767		0		0	0
8/13-19	SS	0	407		0	0	0	0
	DS	0	1,192		0	0	0	0
8/20-26	SS	0	248		1	for August	0	0
	DS	0	1,119		4		0	0
8/27-9/2	SS	0	111		0		0	0
	DS	0	2,625*		1		0	0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
9/3-9	SS	0	TS 3,155*		0		0	0*
	DS	0			0		0	0
9/10-16	SS	0	TS 2,490*		0	0	0	0
	DS	0			0	0	0	1
9/17-23	SS	0	TS 2,644*		0	for September	0	0
	DS	0			0		0	0
9/24-30	SS	0	TS 2,061*		2		0	0
	DS	0			0		0	0*
10/1-7	SS	0	112		1		0	0
	DS	0	1,044		1		0	0
10/8-14	SS	0	115		5	0	0	0
	DS	0	747		2	0	0	0
10/15-21	SS	0	123		4	for October	0	0
	DS	0	1,175*		0		0	0
10/22-28	SS	0	3,175*		2		0	0
	DS	0	417		1		0	0
10/29-11/4	SS	0	215		4		0	1
	DS	0	3,694		4		0	0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at: Loudon, Tenn.	Tennessee River at: Watts Bar Dam	Tennessee River at: Chickamauga Dam
11/5-11	SS	0	68		16		TS 0	TS 0
	DS	0	3,097		6			
11/12-18	SS	0	286		12	0	0	TS 0
	DS	0	4,809		3	0*	1	
11/19-25	SS	0	103		5*	for November	TS 0*	TS 0
	DS	0	3,293		7			
11/26-12/2	SS	0	139		7		0	0
	LS	0	2,230		2		1	1
12/3-9	SS	0	102		0		0	0
	DS	0	3,734		9		1	0
12/10-16	SS	0	130		1	0	0	0
	DS	0	2,208		14	0	2	0
12/17-23	SS	0	53		1	for December	0	0
	DS	0	730		16		0	1
12/24-30	SS	0	45		1		0	0
	DS	0	1,771		6		1	0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
12/31-1/6	SS	0	74		0		0	0
	DS	0	1,699		5		0	0
1/7-13	SS	0	87	0	0		0	0
	DS	0	1,875*	1	6		1	0
1/14-20	SS	0	66	1	0	0	0	0
	DS	1	1,659	9	1	0	0	1
						for January		
1/21-27	SS	0	175	1	1		0	0
	DS	0	1,602	8	20		1	0
1/28-2/3	SS	0	134	1	TS 10*		0	0
	DS	0	773	1			0	0
2/4-10	SS	0	1,584	4	TS 12*		0	0
	DS	0	3,499	5			-1	1
2/11-17	SS	0	371	2	2	0	0	0
	DS	1	2,608	2	5	1	1*	2*
						for February		
2/18-24	SS	0	326	2	1		0	0
	DS	1	1,129	10	4		2	3
2/25-3/3	SS	0	242	1	1		0	0
	DS	0	844	6	3		3	0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
3/4-10	SS	0	173	1	1		0	0
	DS	1	1,216	3	1		0	1
3/11-17	SS	0	TS 1,675*	0	1	0	0	0*
	DS	1		1	0	1	1	0
3/18-24	SS	0	248	-2	1	for March	0*	0
	DS	0	796	0	3		2	2
3/25-31	SS	0	60	2	1		0	0
	DS	1	1,468	6	1		1	1
4/1-7	SS	0	94	0	1		0	0
	DS	1	1,483	13	1		3	2
4/8-14	SS	0	129	3	2	0	0	0*
	DS	0	1,384	3*	13	1	1	1
4/15-21	SS	0	122	2	1	for April	1	0
	DS	1	1,108	7	5		1	1
4/22-28	SS	0	90	1	1		1	0
	DS	3	1,026	1	1		2	1
4/29-5/5	SS	0	0*	0	1		0	0
	DS	2	1,420	1	0		3	1

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
5/6-12	SS	0	150	1	0		0	0
	DS	0	692	1	1		2	0
5/13-19	SS	0	74	1	0	0	0	0
	DS	1	198	1	2	1	0	0
						for May		
5/20-26	SS	0	136	1	1		0	0
	DS	0*	40	2	1		0	1
5/27-6/2	SS	0*	187	0	1		0	0
	DS	0*	423	1	0		1	0
6/3-9	SS	0	436	2	3		0	0
	DS	0	1,931	4	7		0	1
6/10-16	SS	0	584	3	2	0	0	0
	DS	0	2,540	6	5	0	0	0
						for June		
6/17-23	SS	0	258	2	8*		0	0
	DS	0	910	2	3		2	3
6/24-30	SS	0	575	4	6		0	0
	DS	1	1,392	4	6		2	0

*Value is estimated.

SS = suspended solids; DS = dissolved solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
7/1-7	SS	0	534	4	TS 8*		0	0
	DS	1	1,359	4*			1	0
7/8-14	SS	0	392	4	TS 2*	0	0	0
	DS	1	1,822	5		1	3	1
7/15-21	SS	0	173	2	0	for July	0	0
	DS	1	56	0	0		2	0
7/22-28	SS	0*	352	1	0*		0*	0
	DS	5	1,821	4	0		1	0
7/29-8/4	SS	0	310	0	0		0	0
	DS	1	1,482	1	1*		1	1
8/5-11	SS	0	38	TS 20*	TS 16*		0	0
	DS	1	992				0	1
8/12-18	SS	0	196	1	1	0	0	0
	DS	1	97	2	1	1	0	3
8/19-25	SS	0	186	0	0	for August	0	0*
	DS	1	10	0	0		1	1
8/26-9/1	SS	0	141	0	1		0	0*
	DS	1	33	1	1		2	1*

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
9/2-8	SS	0	126	0	0		0	0
	DS	1	17	1	2		0	0
9/9-15	SS	0	177	1	1	0	0	0
	DS	0	774	4	2	1	0	0
9/16-22	SS	0	270	2	3	for September	0	0
	DS	1	2,198	9	10		0	1
9/23-29	SS	0	201	0	1		0	0
	DS	2	1,788	0	4		1	0
9/30-10/6	SS	0	204	1	1		0	0
	DS	0	1,767	5	2		2	0
10/7-13	SS	0	225	2	1		0	0
	DS	1	2,151	6	8		3	0
10/14-20	SS	1	42	1	1	0	0	0
	DS	2	590	2	3	1	1	0
						for October		

SS = suspended solids.

DS = dissolved solids.

Table 3 (Continued)

CONCENTRATIONS OF COBALT-60, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
10/21-27	SS	0*	10	2	0		0	0
	DS	0	1,087	3	2		1	1
10/28-11/3	SS	0	36	2	0		0	0
	DS	2	1,299	2	2		0	0
11/4-10	SS	0	13	2	0		0	0
	DS	1	2,123	8	4		2	1
11/11-17	SS	0	0	2	0	0	0	0*
	DS	0	1,603	14	11	1 for November	2*	0
11/18-24	SS	0*	227	3	1		0	0
	DS	1	1,981	15	15		2	1
11/25-12/1	SS	0	79	2	0		0*	0
	DS	2		6	3		0*	0

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

Even the maximum concentrations of cobalt-60 found at all stations are far below MPC values. Consequently, mean concentrations were not computed.

The distribution of cobalt-60 activity between the suspended and dissolved solids in the samples is summarized in the following tabulation. Percentages are arithmetic averages of all samples. Median percentages are also indicated.

Distribution of Cobalt-60 in Water Samples

Station	Percent Total Activity in			
	Suspended Solids		Dissolved Solids	
	Mean	Median	Mean	Median
Clinch River at Oak Ridge water plant	5	0	95	100
White Oak Creek at White Oak Dam	19	12	81	88
Clinch River at Gallaher Bridge	27	25	73	75
Clinch River at Centers Ferry	30	25	70	75
Tennessee River at Watts Bar Dam	2	0	98	100
Tennessee River at Chickamauga Dam	3	0	97	100

These data indicate 70 to 98 percent of the total cobalt-60 present in the water phase is actually in solution. In White Oak Creek and in Clinch River, approximately 20 to 30 percent of the cobalt-60 present is associated with the suspended solids, but in the Tennessee River the percentage drops to only 2 or 3 percent. This would seem to indicate loss of sediment (and the associated activity) from the water phase in a downriver direction.

Mass Curves--Mass curves of cobalt-60 loads at all stations are shown in figure 9. The curves for White Oak Dam, Gallaher Bridge, and Centers Ferry plot reasonably close together throughout the period of record. Thus there seems to have been no significant loss of this radionuclide in Clinch River. Actually there was an apparent gain in load at Centers Ferry during January and February 1962. However, because of malfunctioning of the sampling equipment at the Centers Ferry station, the reported load at this station might be incorrect. If the curve value for December 1, 1961, is adjusted to about 22 curies (the value obtained by extending the curve established prior to about October 1), and the load thereafter accumulated from this value, the mass curve for this station would fall slightly below the curve for Gallaher Bridge.

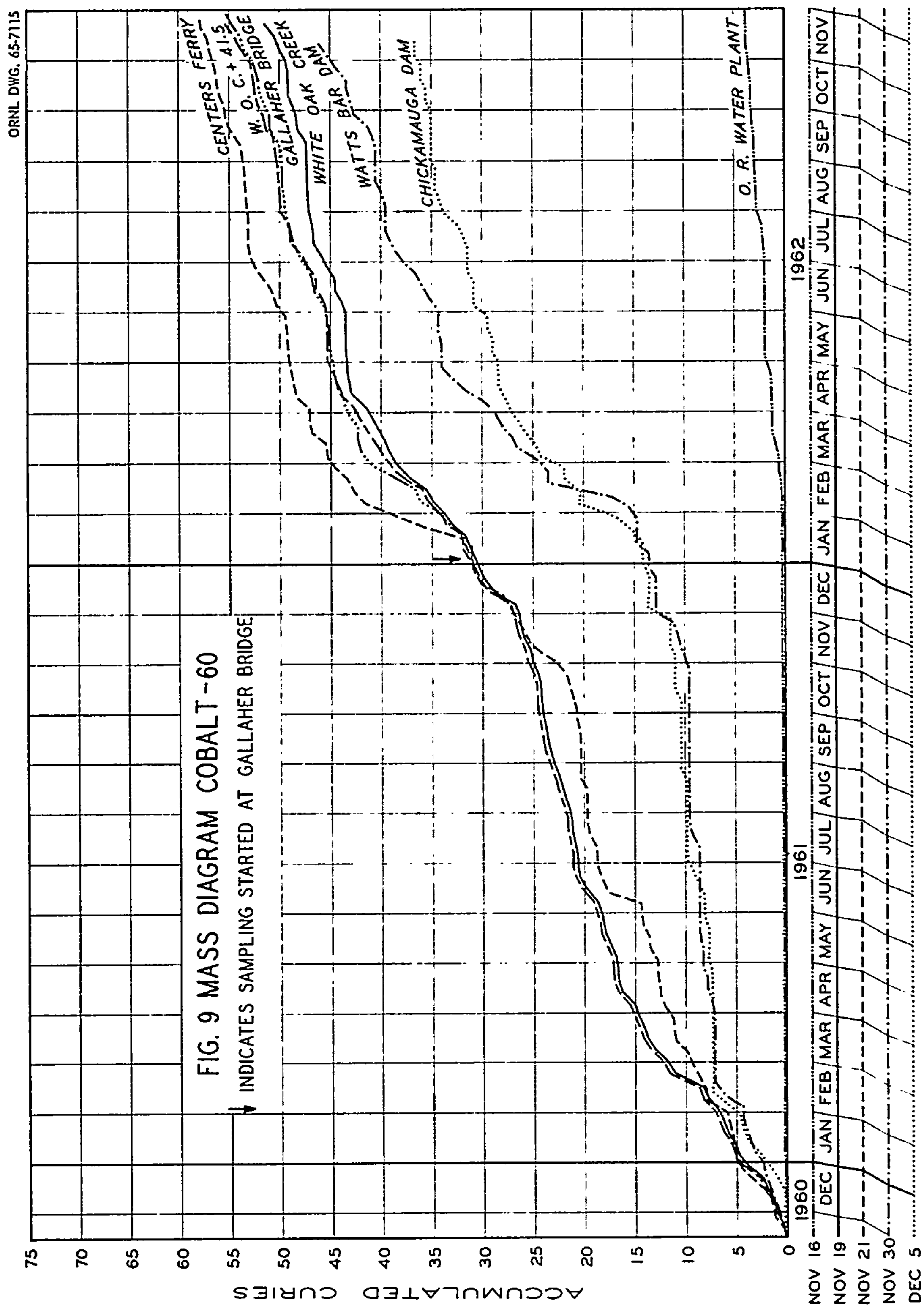

FIG. 9 MASS DIAGRAM COBALT-60

The total load for the two-year sampling period would be about 53 curies. The curves could then be interpreted as showing a very slight loss of cobalt-60 in Clinch River due to sedimentation.

Curves for both Watts Bar and Chickamauga Dams indicate a cumulative loss from the load measured at both White Oak Dam and at Centers Ferry. However, most of this loss is indicated to have occurred during the spring and summer of 1961. From November 1961 through November 1962, the curves for White Oak Creek and Chickamauga Dam are surprisingly parallel. Thus during this period the only effect discernible in the river system was dilution, since the load going in at White Oak Dam arrived later, undiminished, at Chattanooga.

Comparison with Load Measured by ORNL--The data for cobalt-60 plotted in figure 6 indicate serious disagreement in measured loads at White Oak Dam for many of the individual months, but over all the total load during the two-year period was found to be 46.31 curies by ORNL and 51.47 curies by the subcommittee. These values represent a difference of about 10 percent.

Ruthenium-106, Concentrations and Total Stream Loads

Concentrations of ruthenium-106 found in all samples for the period of available record at all stations are shown in table 4.

A factor not noted in earlier progress reports, which could affect reported concentrations to some extent, is the presence of ruthenium-103 (half life = 40 days) and possibly other fission products from weapons fallout, in the samples. Since the mean storage time of the samples prior to counting was approximately 60 to 80 days, measurable quantities of the ruthenium-103 could have been present if the samples contained relatively fresh fallout material. Unfortunately the age and quantities of fallout entering the river cannot be estimated from the available data. Any ruthenium-103 present in the samples would be reported as ruthenium-106 since the respective radionuclides are not distinguishable by the methods used in the study. However, the quantities of ruthenium-103 present are believed to be relatively insignificant in relation to the amounts of ruthenium-106 released via White Oak Creek.

Maximum concentrations found in the weekly (monthly at Loudon) composite samples (including both suspended and dissolved solids) are shown in the tabulation on page 66. Flow-weighted mean concentrations are also shown.

In only White Oak Creek do the maximum concentrations exceed MPC values for drinking water. Mean concentrations at all sampling stations except White Oak Creek at the dam, are far below MPC values for drinking water.

Table 4

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date	Clinch River at Oak Ridge Water Plant		White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
1960								
11/13-19	TS	10	140,424		234			
11/20-26	TS	47	268,169		337			269
11/27-12/3	TS	2	252,368		772		62	38
12/4-10	TS	223	184,714		187		51	74
12/11-17	TS	16	244,604		683	3	77	152
12/18-24	TS	2	192,009		845	for December	59	73
12/25-31	TS	218	217,883		812		117	61
1961								
1/1-7	TS	4	141,791		1,434		96	86
1/8-14	TS	1	171,891		123		174	62
1/15-21	TS	3	294,412		415	0	168	133
1/22-28	TS	1	188,843		384	for January	90	117
1/29-2/4	TS	10	218,938		189		121	88
2/5-11	TS	2	292,517		2,480		100*	97
2/12-18	TS	5	208,479		2,633	2	55	76
2/19-25	TS	7	145,070		415*	for February	112	46
2/26-3/4	TS	10*	87,955		384		192	144
3/5-11	TS	11	98,092		406		91	82
3/12-18	TS	3	125,074		312	SS 0	165	64
3/19-25	TS	2	157,283		347	DS 49	77	38
3/26-4/1	TS	0	144,464		222	for March	66	46

*Value is estimated.

Blank spaces indicate data not available.

TS = total solids; SS = suspended solids; DS = dissolved solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
4/2-8	TS	13	119,620		1,344		61	52
4/9-15	TS	13	106,627		548		56	42
4/16-22	TS	8	121,368		1,263	SS 0	106	38
4/23-29	SS	0*	8,781		117	DS 62	TS 131	TS 46
	DS	123	108,972		260*	for April		
4/30-5/6	SS	14	13,657		39		2	1
	DS	32	15,769		195		84	74
5/7-13	SS	0	9,981		56		2	2
	DS	14	187,918		704		119	30
5/14-20	SS	9*	2,909		32	0	2	8
	DS	20	173,094		392	6	83	107
5/21-27	SS	2	11,004		115	for May	2	5
	DS	6	74,265		523		70	110*
5/28-6/3	SS	1	8,389		91		1	0
	DS	27	76,363		220		83	121
6/4-10	SS	4	76,650*		42		6	4
	DS	12	159,271		215		91	54
6/11-17	SS	3	11,899		219	0	0	2
	DS	11	145,841		646	19	87	61
						for June		

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
6/18-24	SS	4	4,789		304		1	2
	DS	29	123,519		1,154		67	56
6/25-7/1	SS	2	3,386		230		0	1
	DS	7	55,609		740		55	36
7/2-8	SS	24	3,063		3		10	3
	DS	60	67,066		8		120	80
7/9-15	SS	0	3,013		19	1	3	3
	DS	3	88,547		58	3	95	59
						for July		
7/16-22	SS	0	5,841		50		1	3
	DS	7	68,000*		475		64	59
7/23-29	SS	2	1,966		38		3	2
	DS	169	54,769		190		55	29
7/30-8/5	SS	0	3,680		0		6	0
	DS	16	104,740		185		41	36
8/6-12	SS	1	4,756		0		0	0
	DS	0	153,758		148		25	45
8/13-19	SS	1	4,411		0	1	0	0
	DS	185	75,093		5	2	84	18
						for August		

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
8/20-26	SS	1	5,615		13		0	0
	DS	3	90,162		385		20	6
8/27-9/2	SS	9	2,030		7		2	2
	DS	1	180,500*		128		26	39
9/3-9	SS	1	TS 197,800*		2		3	1*
	DS	7			10		41	19
9/10-16	SS	1	TS 154,600*		4	1	1	1
	DS	4			7	64	23	33
						for September		
9/17-23	SS	5	TS 159,750*		5		2	2
	DS	19			6		13	14
9/24-30	SS	1	TS 125,950*		26		0	2
	DS	1			6		7	8*
10/1-7	SS	2	1,331		21		0	1
	DS	5	62,173		26		0	4
10/8-14	SS	1	570		115		2	1
	DS	2	52,204		155		6	3
10/15-21	SS	1	1,076		51	0	1	2
	DS	4	128,700*		42	8	3	1
						for October		

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1961		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
10/22-28	SS	-1	224,100*		27		1	2
	DS	3	21,361		18		17	8
10/29-11/4	SS	2	5,704		58		0	1
	DS	0	209,186		324		10	16
11/5-11	SS	1	1,536		390		TS 5	TS 9
	DS	129	238,627		292			
11/12-18	SS	1	8,313		367	2	1	TS 88
	DS	8	282,698		212	145*	75	
11/19-25	SS	4	2,544		135*	for November	TS 60*	TS 85
	DS	12	199,997		424			
11/26-12/2	SS	4	3,478		320		3	6
	DS	7	138,438		183		54	32
12/3-9	SS	4	3,356		38		6	3
	DS	8	225,385		620		63	42
12/10-16	SS	8	4,576		37	4	0	2
	DS	60	134,500		805	292	73	53
12/17-23	SS	11	1,685		24	for December	5	4
	DS	30	52,830		975		125	80
12/24-30	SS	5	1,298		23		3	6
	DS	24	114,025		404		39	69

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
12/31-1/6	SS	4	1,707		13		14	5
	DS	38	128,816		353		45	33
1/7-13	SS	9	1,913	12	13		6	3
	DS	75	120,000*	182	399		83	53
1/14-20	SS	5	1,585	50	12	8	8	2
	DS	71	108,171	719	376	2	136	210
						for January		
1/21-27	SS	7	3,890	37	24		3	1
	DS	50	103,473	512	542		81	90
1/28-2/3	SS	9	3,026	29	TS 235*		5	5
	DS	3	70,115	213			81	55
2/4-10	SS	6	1,955	21	TS 200*		4	6
	DS	-1	94,595	138			21	42
2/11-17	SS	8	1,960	18	24	8	7	4
	DS	1	93,361	130	121	5	18*	40*
						for February		
2/18-24	SS	24	3,772	52	23		7	8
	DS	8	85,337	482	289		50	53
2/25-3/3	SS	27	2,110	34	20		10	3
	DS	-23	54,187	273	189		104	53

*Value is estimated.

Blank spaces indicate data not available.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
3/4-10	SS	10	2,270	17	21		4	3
	DS	6	79,094	127	60		26	30
3/11-17	SS	11	TS 87,600*	18	10	4	7	4*
	DS	3		73	58	64	57	88
3/18-24	SS	15	3,298	18	25	for March	10*	4
	DS	4	44,402	80	171		25	33
3/25-31	SS	10	1,259	52	13		13	6
	DS	-1	85,423	238	174		52	23
4/1-7	SS	12	1,772	14	41		9	9
	DS	9	81,578	635	97		30	33
4/8-14	SS	31	3,320	86	64		6	12*
	DS	34	72,501	200*	680		41	32
4/15-21	SS	6	1,971	78	42	1	6	7
	DS	-3	52,418	284	376	32	75	47
4/22-28	SS	5	1,698	24	16	for April	15	7
	DS	27	62,902	87	78		51	61
4/29-5/5	SS	4	1,700*	13	11		1	5
	DS	41	81,897	66	40		65	41

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
5/6-12	SS	3	1,900	22	11		3	5
	DS	-2	40,756	53	36		26	22
5/13-19	SS	4	882	17	13	1	2	3
	DS	3	13,735	22	45	2	33	4
5/20-26	SS	4	817	12	10	for May	4	1
	DS	4*	36,860	27	14		18	8
5/27-6/2	SS	TS 5*	2,153	17	12		2	4
	DS		23,237	23	14		9	11
6/3-9	SS	3	7,850	31	53		4	5
	DS	-1	108,784	140	276		11	9
6/10-16	SS	5	8,553	52	42	5	0	0
	DS	2	132,388	215	342	8	7	4
6/17-23	SS	7	3,982	39	320*	for June	1	1
	DS	-1	56,020	72	148		7	1
6/24-30	SS	4	17,146	69	110		1	2
	DS	-1	46,056	204	247		16	12

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
7/1-7	SS	1	13,168	83	TS 250*		2	4
	DS	-1	64,164	170*			21	16
7/8-14	SS	3	6,397	88	TS 225*		3	1
	DS	0	92,971	223			21	13
7/15-21	SS	2	790	25	130*	2	5	1
	DS	3	3,260	32	15	3	30	18
7/22-28	SS	0*	3,538	22	100*	for July	2*	1
	DS	15	91,380	118	28		25	15
7/29-8/4	SS	3	3,714	9	TS 80*		2	3
	DS	8	74,957	78			17	33
8/5-11	SS	5	1,811	TS 98*	TS 65*		2	3
	DS	5	46,233				13	28
8/12-18	SS	2	665	17	24	0	4	1
	DS	4	3,111	65	-5	4	-5	19
8/19-25	SS	4	1,105	6	4	for August	1	0*
	DS	9	1,206	4	14		18	18

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
8/26-9/1	SS	2	350	3	3		2	0*
	DS	7	1,107	4	8		8	7*
9/2-8	SS	4	338	5	5		1	0
	DS	0	1,127	21	5		0	1
9/9-15	SS	3	1,800	11	9	1	1	2
	DS	-1	34,130	91	54	6	12	2
						for September		
9/16-22	SS	4	3,740	39	70		1	1
	DS	21	89,381	363	436		11	0
9/23-29	SS	4	1,870	8	10		2	1
	DS	3	77,720	101	136		8	8
9/30-10/6	SS	4	2,798	21	17		3	1
	DS	13	61,193	168	103		33	0
10/7-13	SS	4	2,644	27	30	0	4	5
	DS	6	72,965	197	298	3	22	4
						for October		

*Value is estimated.

SS = suspended solids.

DS = dissolved solids.

Table 4 (Continued)

CONCENTRATIONS OF RUTHENIUM-106, pc per liter

Date 1962		Clinch River at Oak Ridge Water Plant	White Oak Creek at Dam	Clinch River at Gallaher Bridge	Clinch R. above Centers Ferry	Tennessee River at Loudon, Tenn.	Tennessee River at Watts Bar Dam	Tennessee River at Chickamauga Dam
10/14-20	SS	12	524	23	20		4	2
	DS	5	22,192	38	75		25	21
10/21-27	SS	4*	440	20	7		2	4
	DS	11	38,291	53	41		75	32
10/28-11/3	SS	0	940	25	7		1	2
	DS	8	49,971	46	63		23	23
11/4-10	SS	6	1,058	47	11	5	6	3
	DS	2	73,832	247	155	6 for November	15	11
11/11-17	SS	10	417	74	20		2	2*
	DS	7	60,470	462	391		23*	5
11/18-24	SS	0*	4,035	57	24		3	2
	DS	17	67,577	655	686		25	6
11/25-12/1	SS	6	1,388	43	13		TS 30*	4
	DS	7		149	129			26

*Value is estimated.

SS = suspended solids; DS = dissolved solids; TS = total solids.

Maximum and Mean Concentrations of Ruthenium-106

Station	Highest Concentration	Period of Occurrence	Flow-Weighted Mean Concentration
	pc per liter		pc per liter
Clinch R. at Oak Ridge water plant	223	Dec. 4-10, 1960	23
White Oak Creek at White Oak Dam	294,412	Jan 15-21, 1961	109,800
Clinch R. at Gallaher Bridge	769	Jan. 14-20, 1962	345*
Clinch R. at Centers Ferry	2,633	Feb. 12-18, 1961	317
Tenn. R. at Loudon, Tenn.	296	December 1961	**
Tenn. R. at Watts Bar Dam	192	Feb. 26-Mar. 4, 1961	63
Tenn. R. at Chickamauga Dam	269	Nov. 20-26, 1960	51

*Record begun January 8, 1962.

**Not applicable.

The distribution of ruthenium-106 activity between the suspended and dissolved solids in the samples is summarized in the following tabulation. Percentages are arithmetic averages of all samples. Median percentages are also indicated.

Distribution of Ruthenium-106 in Water Samples

Station	Percent Total Activity in			
	Suspended Solids		Dissolved Solids	
	Mean	Median	Mean	Median
Clinch River at Oak Ridge water plant	44	29	56	71
White Oak Creek at White Oak Dam	6	4	94	96
Clinch River at Gallaher Bridge	21	17	79	83
Clinch River at Centers Ferry	21	16	79	84
Tennessee River at Watts Bar Dam	11	7	89	93
Tennessee River at Chickamauga Dam	15	8	85	92

From these data it is apparent that from 79 to 94 percent of the ruthenium-106 activity is associated with the dissolved solids, or in other words, dissolved in the water itself. Neither the time of contact with the suspended solids, nor sedimentation, appear to have any significant influence on the distribution of activity between suspended solids and dissolved solids since the percentage associated with the dissolved solids decreases from 94 percent at White Oak Dam to 79 percent at Centers Ferry, then goes back up to 89 percent at Watts Bar, and back down to 85 percent at Chickamauga Dam.

Mass Curves--Cumulative curves of ruthenium-106 loads at all stations except Loudon are shown in figure 10.

During the first year of sampling, the mass curves for all stations below White Oak Creek agree quite closely with that for White Oak Creek; then, beginning in the fall of 1961 and continuing through March 1962, the downriver curves diverge to some extent. From March through November 1962, the curves remain essentially parallel to each other. The divergence in early 1962 appears to reflect the effect of fallout from weapons testing. (See figure 5, page 23.)

Throughout the two-year period the curve for Centers Ferry is practically identical to the one for White Oak Creek. Likewise, during the last 11 months of record the curve for White Oak Creek plus the Oak Ridge water plant is nearly identical to that for Gallaher Bridge. Throughout the entire period of record, the curves for Watts Bar Dam and Chickamauga Dam are essentially the same.

Based on the rather amazing agreement between the cumulative loads measured at all stations below White Oak Dam with the load measured at White Oak Dam, it can be definitely concluded that during the two-year sampling period essentially all the ruthenium-106 discharged from Oak Ridge passed through the river system to Chattanooga in the water phase. That ruthenium-106 which is found in bottom sediments between Oak Ridge and Chattanooga must represent the continued accumulation over the years of a very small percentage of the annual load discharged at Oak Ridge.

The good agreement in the cumulative loads measured at the successive stations indicates considerable confidence can be placed in the methods used throughout the study in sampling and compositing.

Comparison with Loads Measured by ORNL--As with the other radionuclides, comparison of monthly loads at White Oak Dam as measured by the subcommittee and by ORNL, indicates several rather serious disagreements, as shown in figure 7, page 25. However, comparison of cumulative loads measured over longer periods indicates better agreement, as might be expected.

The first 12-month period (December 1960 through November 1961) shows a total discharge of about 1,900 curies of ruthenium-106 at White Oak Dam as measured by the subcommittee, while the second 12-month period

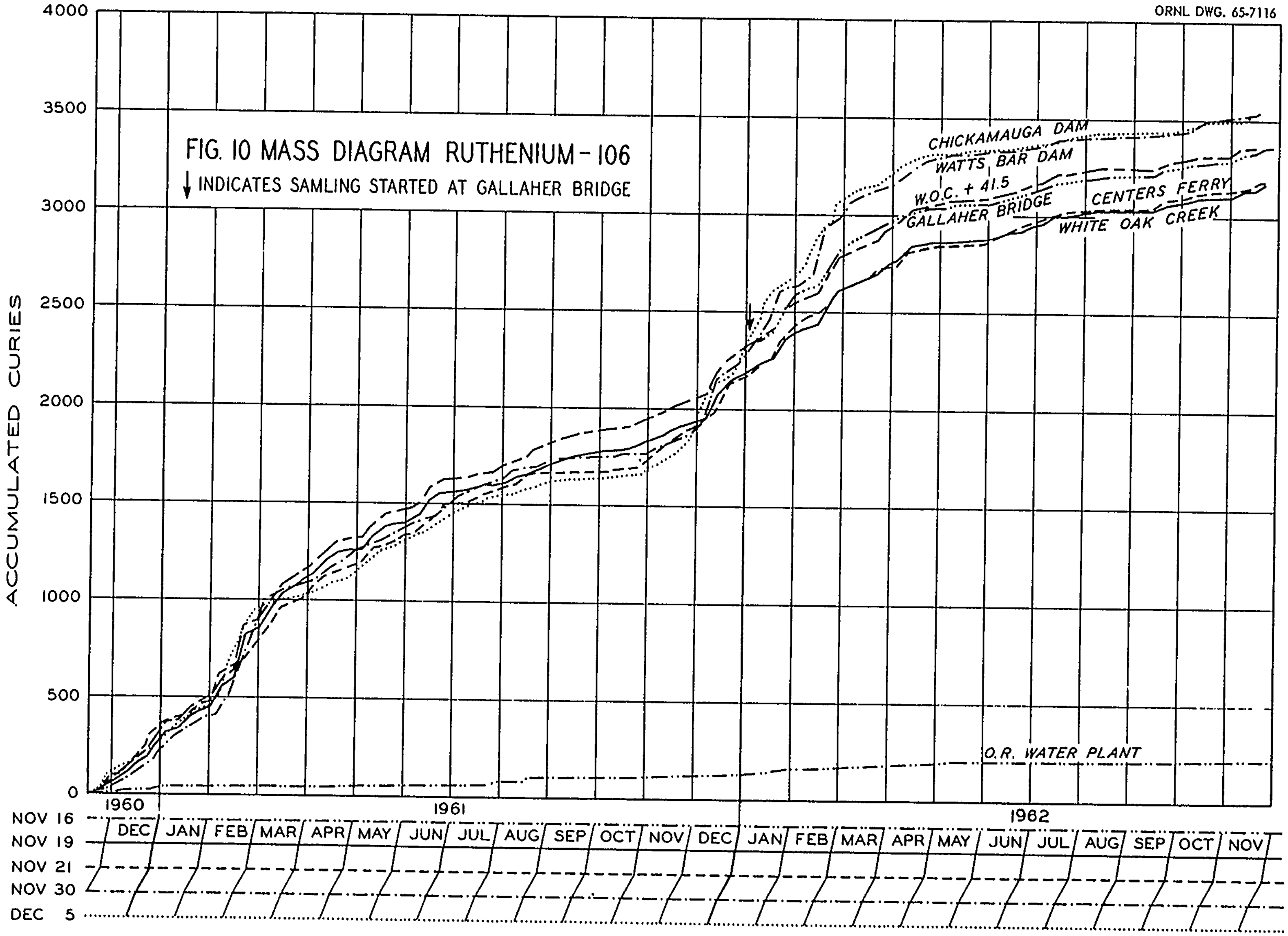

FIG. 10 MASS DIAGRAM RUTHENIUM-106

↓ INDICATES SAMLING STARTED AT GALLAHER BRIDGE

(December 1961 through November 1962) shows a total discharge of about 1,300 curies. Data supplied by the Oak Ridge National Laboratory show a total discharge of 1,906 curies at White Oak Dam during the first 12-month period and a total discharge of 1,586 curies for the second 12-month period. Thus in comparing the total loads reported in this study with those reported by ORNL, the total loads for the first 12-month period are found to be identical. On the other hand, the difference of nearly 300 curies during the last 12-month period represents a disagreement of approximately 22 percent. If the White Oak Creek load for the last 12 months of record as measured by the subcommittee, were increased by 300 curies, it would equal almost exactly that measured for Chickamauga Dam. This probably indicates a negative bias in the White Oak Creek values reported by the subcommittee for the last 12-month period. (See figure 10, page 68.)

Effects of Operation of Melton Hill Dam on Dispersion of Radionuclides

The operation of Melton Hill Dam at mile 23.1 on Clinch River will change the hydraulic pattern of releases of radioactive waters from White Oak Creek into Clinch River. The potential effects of this altered hydraulic pattern on the dispersion of radioactive waters originating at the Oak Ridge National Laboratory have been investigated cooperatively in the field by personnel of the U. S. Geological Survey and of the Oak Ridge National Laboratory.

From results of these dispersion studies that have been and are being reported separately, the subcommittee concludes that although the time versus concentration pattern of radionuclides will be altered drastically in the Clinch River embayment of Watts Bar Reservoir, the pattern of dispersion in the Tennessee River will not be altered sufficiently from that observed during the two-year study reported here to justify reactivation of the network of sampling stations.

Recommendations

Recommendations 1, 2, and 3 concern the monitoring program which the subcommittee feels must be continued indefinitely at and below Oak Ridge. Recommendations 4, 5, and 6 concern improvements that should be made in studies of the type reported here, should such a study be reinitiated in the future at and below Oak Ridge, or put into operation by others at some other location for similar purposes.

1. (a) Continuous monitoring and proportional sampling should be continued at White Oak Dam, and weekly composite samples should be examined for concentrations of strontium-90, cesium-137, cobalt-60, and ruthenium-106. Arrangements should be made to keep this station rated since knowledge of continuous streamflow rates at this station is essential.

(b) Proportional sampling should be initiated very soon and continued indefinitely on the power discharge of Melton Hill Dam. Weekly composite samples should be examined for radionuclide activity.

(c) Proportional sampling should be initiated very soon and continued indefinitely at or near the present location of the water intake in Clinch River of the Oak Ridge Gaseous Diffusion Plant. Volumes of river water, proportioned to the instantaneous rate of river discharge at the intake site, should be added to the composite sample at intervals of not more than 15 minutes. Such samples, composited weekly, should be examined for the radionuclides of importance unless sample results at White Oak Dam, or at Melton Hill Dam, indicate need for more frequent examination.

(d) If at any time in the future it becomes reasonably possible for any significant load of radionuclides to enter Clinch River downstream from the monitoring station at the water intake of the Oak Ridge Gaseous Diffusion Plant, either the station should be moved far enough downstream to intercept such additional inflow, or an additional downstream monitoring station should be established.

2. Since the Oak Ridge National Laboratory and the Public Health Service will both be monitoring Clinch River below White Oak Creek, these two agencies, and any others that may collect radiological samples here, should compare results obtained on regularly scheduled split samples. This is essential to prevent differences in technique, equipment, etc., from introducing disagreement in routine sampling results.

3. The Public Health Service should be supplied with part of each weekly composite sample collected at the water intake of the Oak Ridge Gaseous Diffusion Plant. The Public Health Service should be requested in a letter from the Chairman of the Clinch River Study Steering Committee, to utilize these samples in the radiological determinations made on water samples collected at this station in its Water Pollution Surveillance System.

4. If any detailed study of this nature is made in the future, it would be extremely helpful in determining cesium-137 activity levels if this radionuclide were extracted from the sample by the best chemical separation technique available, prior to counting. In any situation where the gamma spectrum of a radionuclide of importance is seriously masked by some other radionuclide, chemical separation, as well as gamma spectrometry, should be used. The most sensitive, yet accurate, technique and

equipment available should be applied to the determination of the concentration of each radionuclide. It would be helpful if a few "dry runs" were made at all proposed sampling stations prior to the initiation of routine sampling, to determine the volumes of samples needed at the various stations to provide sufficient activity, after concentration, for accurate measurements.

5. It is recommended for any future study of this nature that the actual, and relative concentrations of radionuclides in the suspended and dissolved solids be determined as carefully as possible in a limited number of special samples collected at such times as would permit detection of the influences, if any, on relative concentrations, of such factors as water chemistry, streamflows, particle sizes, time of flow below Oak Ridge, water temperature, and possibly other variables. To provide needed information on precision and reproducibility of results, more effort than in the present study should be directed toward duplicate processing of " whole samples," and in processing duplicate samples (twice the needed volume, mixed and split).

6. If a network of sampling stations is needed for future studies of this general nature, a companion network of precipitation stations would be desirable to provide information on the fallout contribution to the radionuclide load.

Respectfully submitted,

SUBCOMMITTEE ON WATER SAMPLING AND ANALYSIS

M A Churchill

M. A. Churchill, Chairman
J. S. Cragwall, Jr.
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RADIOACTIVE MATERIALS IN BOTTOM SEDIMENT OF CLINCH RIVER: PART A, INVESTIGATIONS OF RADIONUCLIDES IN UPPER PORTION OF SEDIMENT

P. H. Carrigan, Jr.
R. J. Pickering
T. Tamura
R. Forbes

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HEALTH PHYSICS DIVISION

RADIOACTIVE MATERIALS IN BOTTOM SEDIMENT OF CLINCH RIVER: PART A, INVESTIGATIONS OF RADIONUCLIDES IN UPPER PORTION OF SEDIMENT

Supplement No. 2A to Status Report No. 5 on Clinch River Study

By

P. H. Carrigan, Jr. (USGS)
R. J. Pickering (USGS)
T. Tamura (ORNL)
R. Forbes (TVA)

Final Progress Report, Part A, of Subcommittee on Bottom Sediment Sampling and Analysis

MARCH 1967

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

ACKNOWLEDGMENT

The Subcommittee on Bottom Sediment Sampling and Analysis arranged for canvass of information pertaining to the association of radionuclides with bed deposits of the Clinch and Tennessee Rivers. From this compilation, a summary of available data pertinent to the area of study of the subcommittee[1] was prepared.

The Subcommittee on Bottom Sediment Sampling and Analysis was appointed in May 1961 to help coordinate and implement the investigations of the Clinch River Study[2].

Membership of the subcommittee has been as follows: P. H. Carrigan, Jr., (USGS), Chairman (1961-3, 1965); R. M. Richardson (USGS), Chairman (1964); H. H. Abee (ORNL); T. Tamura (ORNL); J. Smallshaw (TVA, 1961-3, ret.); J. W. Beverage (TVA, 1963); and R. W. Andrew (USPHS). Staff to the subcommittee has been as follows: W. P. Bonner (ORNL); R. Forbes (TVA); R. J. Pickering (USGS); and S. L. Cummings (USPHS).

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Parts of the text of this report have appeared as short progress reports of investigations in Status Reports Nos. 2-5 on Clinch River Study[1-4], and a summary of this report is in Status Report No. 5 on Clinch River Study[4].

INTRODUCTION

Radionuclides in the bottom sediment of the Clinch and Tennessee Rivers originate from three sources: naturally occurring radioactive materials in the earth, fallout from weapons testing, and the release of low-level radioactive waste from various industrial and research facilities to streams in eastern Tennessee. The principal source is the Oak Ridge National Laboratory (ORNL). Operations began at the Oak Ridge National Laboratory in 1943.

Radioactive waste effluents from facilities at ORNL pass through a waste processing system. The processed liquid wastes, containing low-level radioactivity, enter the Clinch River via White Oak Creek (Fig. 1).

Health physicists at ORNL noted radioactive materials associated with bottom sediments in the creek embayment (0.6 mile long) downstream from White Oak Dam. Later (1951) their investigations indicated radioactivity in bottom sediment of the Clinch and Tennessee Rivers.

Origin of Fission Product Wastes

Fission products are discharged into White Oak Creek basin from the process waste water treatment system, waste seepage pits, and several minor sources not included in the main waste systems at ORNL[9]. Intermediate-level radioactive liquid wastes (more than 1.0 microcurie per gallon) flow in special drains to storage tanks centrally located in the ORNL plant area. The wastes are transferred by pipeline to waste seepage pits. Low-level radioactive waters seep from these pits into White Oak Creek. Low-level radioactive liquid wastes (less than 1.0 microcurie per gallon) flow in another sewage system to the process waste water treatment area. After treatment these waste waters are released into White Oak Creek.

Some facilities at ORNL are not connected into either the intermediate-level or low-level radioactive liquid waste sewage networks. Drainage of low-level radioactive materials from these facilities into White Oak Creek or tributaries is monitored to insure against discharge of waters containing radioactivity above prescribed limits.

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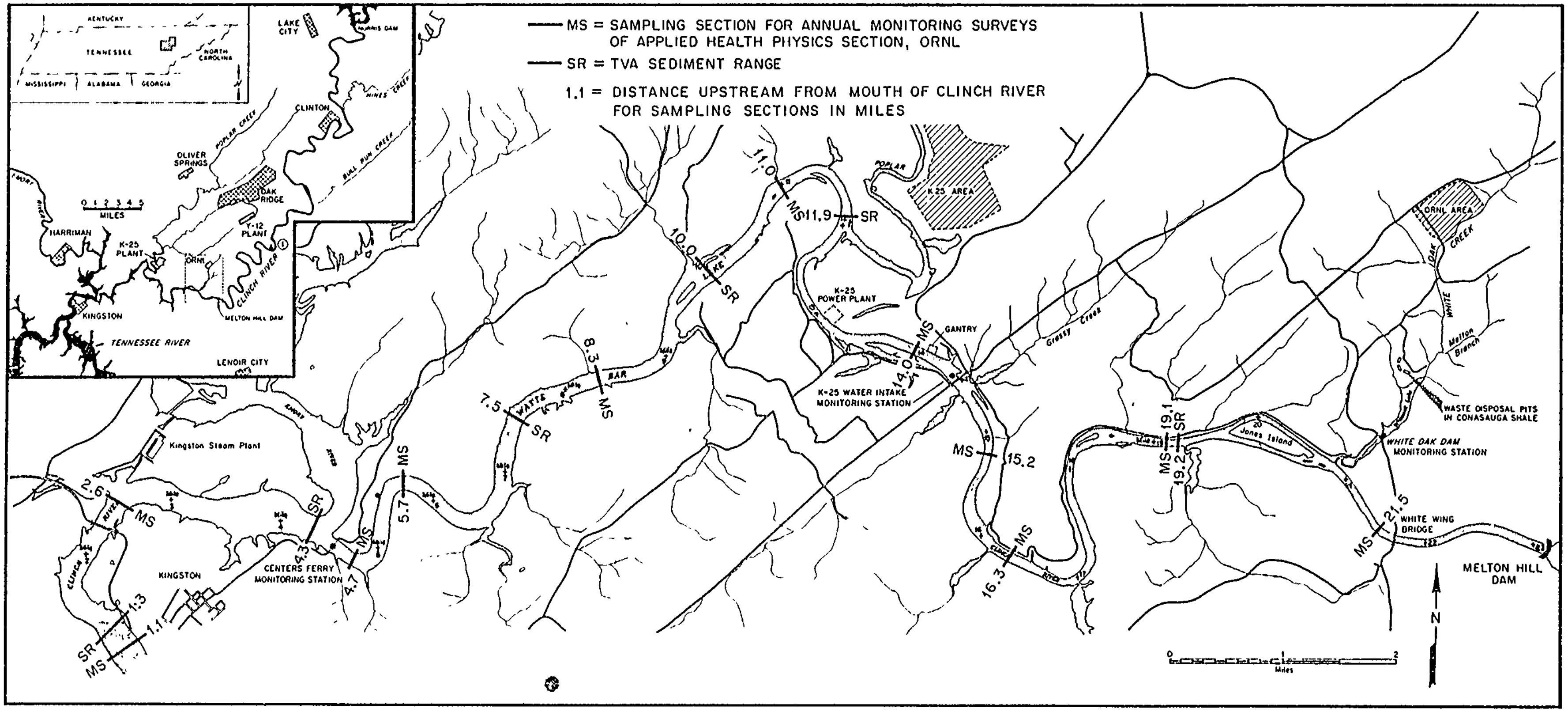

Fig. 1. Map of Clinch River Showing Bottom Sediment Observation Sections.

Objective of Report

Sources of information pertaining to the various investigations concerned with radioactivity in the bed sediments are diverse; some information is in the open literature, some in reports issued by ORNL, and much is in the files of the investigators and of the various agencies involved in the Clinch River Study. In this report information about radionuclides in bottom sediment of the Clinch and Tennessee Rivers, pertinent to objectives of the Clinch River Study is summarized and interpreted.

The objectives of the Study, as set forth by the Clinch River Study Steering Committee[10], are

(1) To determine the fate of radioactive materials currently being discharged to the Clinch River,

(2) To determine and understand the mechanisms of dispersion of radionuclides released to the river,

(3) To evaluate the direct and indirect hazards of current disposal practices in the river,

(4) To evaluate the over-all usefulness of this river for radioactive waste disposal purposes, and

(5) To provide appropriate conclusions regarding long-term monitoring procedures.

Two factors tend to define the scope of information in this report. First, most information is from investigations in progress or completed at the time of canvass of available information, June 1962. Second, a substantial change in flow conditions has occurred in the Clinch River downstream from Melton Hill Dam since this structure was completed (Fig. 1). Because these changes in flow conditions may affect the movement of radionuclides in the river, only investigations conducted prior to the time operations began at Melton Hill Dam are included in the report.

Two important studies pertaining to the Study have not been fully treated in this report. These studies are: (1) a firm estimate of the quantity of fission products in the bed of the Clinch River and (2) a description of the vertical distribution of radioactivity in bottom sediments of the Clinch and of geochemical factors which influence this distribution. These studies are the subject of Supplement 2B.[4]

Available Data

At five-year intervals, beginning in 1946, personnel of the Hydraulic Data Branch, TVA, measured the elevation of the surface of the bottom sediments of the Clinch River at selected sections. The purpose of these measurements was (1) to estimate the number of years that sedimentation will occur (a) before sedimentation interferes with water-control operations in Watts Bar Lake and (b) before the useful life of the reservoir is ended, and (2) to determine the current capacity of the reservoir[11]. These sections, called sediment ranges, are located at Clinch River Miles (CRM) 1.3 (1.3 miles upstream from mouth of river), 4.3, 7.5, 10.0, 11.9, and 19.2 (Fig. 1). Similar work has been done in other reservoirs on the Tennessee River.

Each year, beginning in 1951, personnel of the Applied Health Physics Section, ORNL, made surveys of the radioactivity in the bottom sediments of the Clinch and Tennessee Rivers. From these surveys, they evaluated potential, present, and future hazards resulting from radioactivity in these sediments, predicted the capacity of the sediments for storing radioactive materials, recommended rates of release of radioactive wastes to the Clinch River, and determined the effect of increased radioactivity in sediments in the river system on industry[6]. The surveys extended from Norris Lake on Clinch River anf from Fort Loudoun Lake on Tennessee River into Guntersville Lake on Tennessee River (in 1952 and 1961 Kentucky Lake included; see Figs. 1 and 2)[5-8].

In 1960, personnel of the Waste Disposal Research Section, ORNL, collected core samples of bottom sediments in 19 sections of the Clinch River, from CRM 4.7 to 22.5. L. Hemphill and W. B. Nix of ORNL and P. H. Carrigan, Jr., of USGS used the cores for study of the distribution of radionuclides in the upper horizon of Clinch River bottom sediments. T. Tamura, ORNL, determined physicochemical characteristics and sorption capacity of the composites of the cores.

Composites of cores which were collected in 1961 at five sections in the Clinch River, from CRM 4.7 to 19.2, were sent to USGS

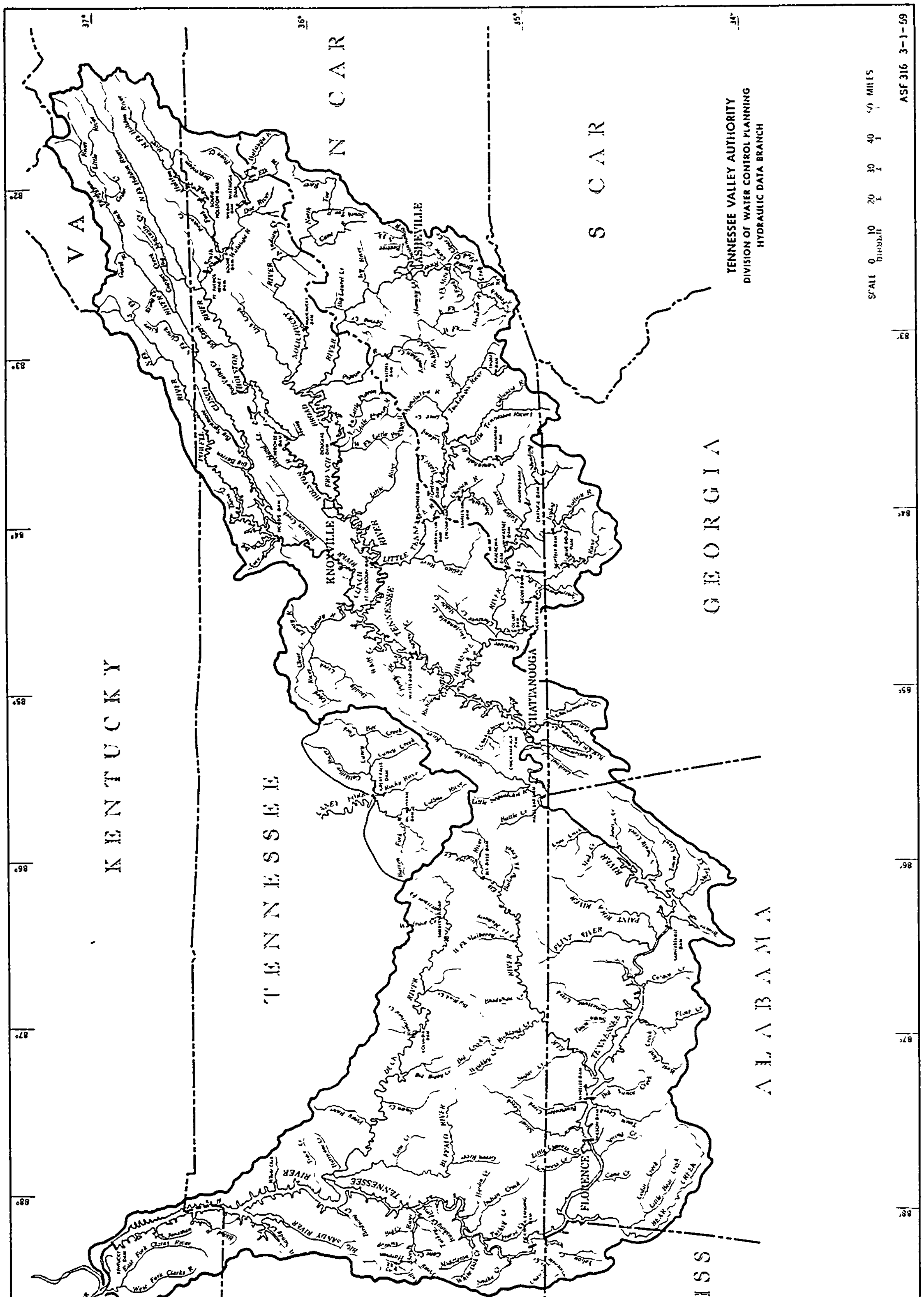

Fig. 2. Tennessee River Basin

laboratories (Denver, Colorado, and Releigh, North Carolina) for mineralogical, cation exchange, and size-distribution analyses. Study of these analyses was the first step in describing the physicochemical properties of the cores.

Scientists of the Division of Radiologic Health, USPHS, Cincinnati, Ohio, undertook investigations of bottom sediments of the Clinch and Tennessee Rivers as well. They studied the variation of radionuclide concentration with particle size and with location in the study reaches, particularly for bottom sediment of the Tennessee River.

Information on flows in the Clinch and Tennessee Rivers used in this report has been obtained from USGS and TVA reports.

DESCRIPTION OF STUDY REACH

Studies of radionuclides in river-bottom sediments have extended from Norris Lake on the Clinch River and Fort Loudoun Lake on the Tennessee River as far downstream as Kentucky Lake. A map showing the portion of the Tennessee River System included in these studies appears in Fig. 2.

The Clinch River enters the Tennessee River in the backwater pool of Watts Bar Lake, and contributes the minor portion of the inflow to the lake. Drainage area of the Clinch River at Norris Dam is 2,912 square miles, and at the mouth it is 4,413 square miles. Drainage area of the Tennessee River at Fort Loudoun Dam is 9,550 square miles, which is increased by the Little Tennessee River and minor drainage to 12,470 square miles just upstream from the mouth of the Clinch River.

The Clinch River portion of the Tennessee River basin lies principally in the Valley and Ridge Province. Sediment flowing into the study reaches is largely that contributed by minor tributaries of the Valley and Ridge Province, because the major storage reservoirs--Norris on the Clinch River, Cherokee on the Holston River, Douglas on the French Broad River, and Fontana and three downstream power pools on the Little Tennessee River--trap most of the sediment from the upper 12,850 square miles (76 percent) of the drainage area above the mouth of the Clinch River. As the sediment load passes downstream, a succession of reservoirs (Table 1) acts to retard the flow and accumulate bottom sediment from the suspended material.

For study reaches in the Clinch and Tennessee River, water levels are affected by reservoir impoundments, and flow is affected by water-control operations. In the Clinch River, backwater of Watts Bar Lake extended as far upstream as CRM 25 in the winter and CRM 28 in the summer prior to the closure of Melton Hill Dam.

Thermal stratification affects flow through these reservoirs[12,13]. During summer months, cold water from Norris Lake begins to underflow the warm, stilled water of the lower Clinch River embayment in the vicinity of CRM 12. Practical use has been made of this condition by construction of a submerged dam at CRM 3.8, just below the Emory

Table 1. Location of Clinch River Tributaries and Tennessee River Dams[a]

Identification	Distance Above Mouth (miles)	Distance Downstream from White Oak Creek (miles)
	Clinch River	
Mouth of White Oak Creek	CRM 20.8	0.0
Mouth of Poplar Creek	12.0	8.8
Mouth of Emory River	4.4	16.4
Submerged dam for underflow diversion	3.8	17.0
	Tennessee River	
Mouth of Clinch River	TRM 567.7	20.8
Watts Bar Dam	529.9	58.6
Chickamauga Dam	471.0	117.5
Hales Bar Dam	431.1	157.4
Guntersville Dam	349.0	239.5
Wheeler Dam	274.9	313.6
Wilson Dam	259.4	329.1
Pickwick Landing Dam	206.7	381.8
Kentucky Dam	22.4	566.1

[a]Dams on the Clinch River upstream from White Oak Creek are Norris at CRM 79.8 and Melton Hill at CRM 23.1. The mean annual flow of the Clinch River near Scarboro, Tennessee, was 4,564 cubic feet per second (cfs) for the 24-year period 1936–60. Following are the long-term mean annual flows at principal stations on the Tennessee River for the periods indicated:

Station	Period of Record (years)	Mean Annual Flow (cfs)
Knoxville, Tennessee	61	12,810
Chattanooga, Tennessee	86	37,030
Florence, Alabama	66	50,620
Paducah, Kentucky	71	63,790

River mouth, to divert this cold water up the Emory River to the condenser intakes of the Kingston Power Plant. The condenser effluent returns to the lower Clinch River.

In the future, thermal differences, resulting from operation of Bull Run Power Plant (under construction at CRM 47.6), will stratify flow to an appreciable degree in Melton Hill Lake. The first unit of the plant will have a capacity of 900 megawatts. The cooling water will be pumped through the unit at 928 cubic feet per second and will be heated about 18 degrees Fahrenheit.

SEDIMENT DEPOSITION

Whether retention of fission products in bottom sediments of the Tennessee River basin is a major factor in determining the fate of the releases or constitutes a potential hazard, depends on the concentrations of the fission products in the bottom sediment, mass of sediment accumulated on the stream beds, and movement of the deposits subsequent to deposition. In later sections of the report, concentrations of the fission products in the sediments will be considered. In this section, the location and extent of the sediment deposits will be presented.

Sediment deposition is considered only in Watts Bar and Chickamauga Lakes (see Fig. 2). Consideration of sediment deposition elsewhere is not included because radionuclide concentrations in the sediment are extremely low.

Description of variations in sediment volume for the period of record comes from TVA sediment range surveys. See Figs. 1, 3, and 4 for locations of sediment ranges, and Fig. 5 and Tables 2 and 3 for variations in volume of bottom sediment in study reaches.

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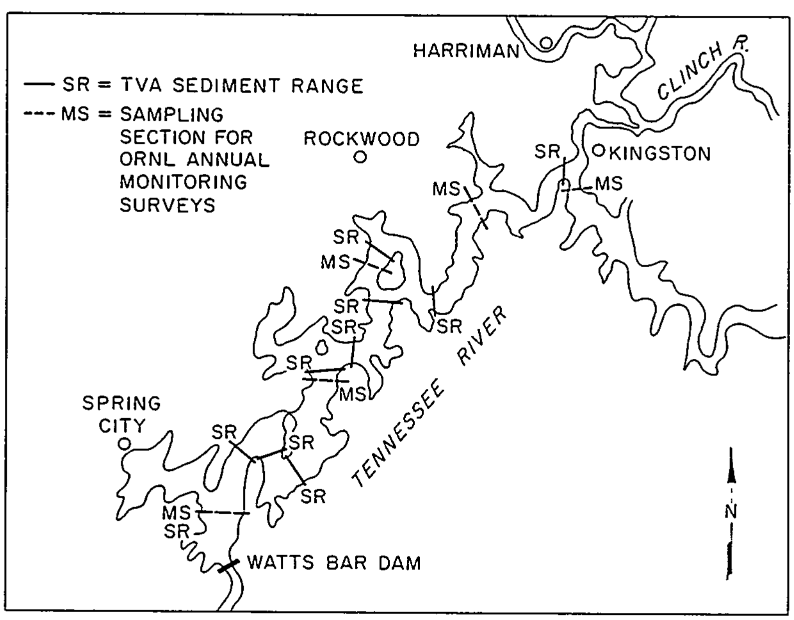

Fig. 3. Map of Watts Bar Lake Showing Bottom Sediment Observation Sections

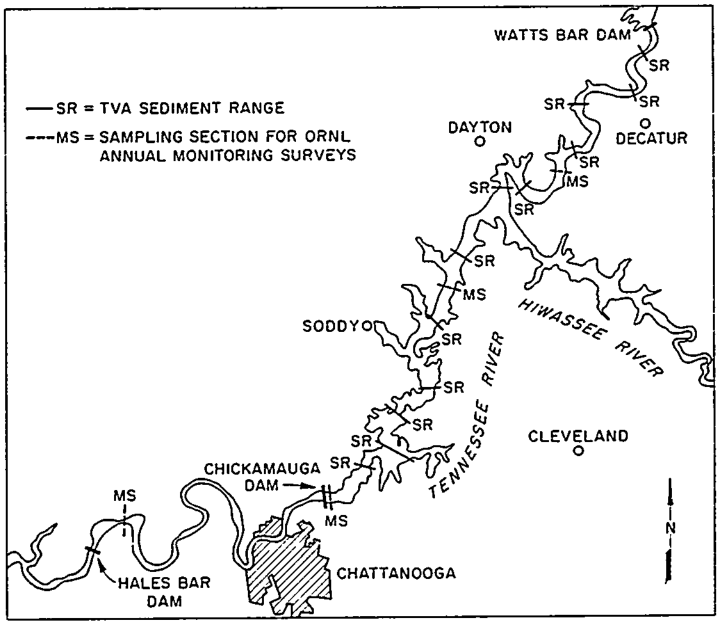

Fig. 4. Map of Chickamauga Lake Showing Bottom Sediment Observation Sections

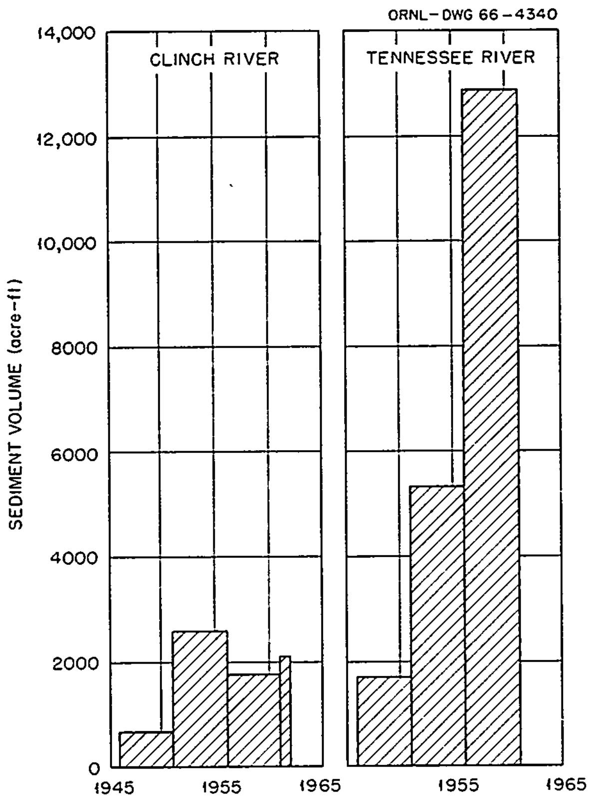

Fig. 5. Total Accumulation of Bottom Sediments in Clinch and Tennessee River Embayments of Watts Bar Lake, 1946 to 1962, and to 1961, Respectively.

Table 2. Change in Volume of Bottom Sediment in Tennessee River Embayment of Watts Bar Lake for Indicated Periods

Reach (TRM)		Change in Volume (acre-feet per mile)		
Begin	End	1946–51	1951–56	1956–61
529.9	532.1	98	–55	278
532.1	534.7	85	36	327
534.7	537.8	47	179	226
537.8	538.8	134	347	346
538.8	543.7	40	158	142
543.7	546.2	14.7	77	200
546.2	549.9	11.0	–30	210
549.9	552.7	44	25	241
552.7	557.1	44	147	181
557.1	562.2	34	102	174
562.2	567.7	22	90	96

Table 3. Change in Volume of Bottom Sediment in Tennessee River Embayment of Chickamauga Lake for Indicated Periods

Reach (TRM)		Change in Volume (acre-feet per mile)			
Begin	End	1940–47	1947–54	1954–56	1956–61
471.0	475.1	211	–89.3	89.3	–2.46
475.1	478.2	180	3.00	–90.7	73.6
478.2	480.8	534	–338	–153	237
480.8	484.8	243	–209	–62.1	157
484.8	490.4	271	–190	55.2	80.0
490.4	496.4	290	–274	166	149
496.4	501.7	236	–230	184	112
501.7	506.6	156	–119	97.0	–32.4
506.6	514.3	142	–176	94.7	–34.4
514.3	517.9	82.5	–104	6.41	6.28
517.9	523.2	88.7	–89.4	–30.1	2.62
523.2	527.3	89.3	–108	19.8	–32.5
527.3	529.9	29.3	–45.5	12.9	0.29

In the Clinch River downstream from the Emory River, the rate of accumulation of bottom sediments averaged a nearly steady 931 acre-feet per year (1946-62). Upstream from the Emory River, alternate periods of gains and losses in volume occurred, with gains and losses being nearly equal over the period of record from CRM 4.3 to 7.5 and from CRM 11.9 to 19.2. The continuous and uniform accumulation of bed material downstream from Emory River, in contrast to the alternate periods of gains and losses upstream from this section, appears to result from extremely low velocities, and deposition of sediments contributed from the Emory River basin. Larger changes in volume of bottom sediments observed in the reach between CRM 7.5 and 11.9 than in the reaches immediately upstream or downstream, seem to result from loads discharged from Poplar Creek (at CRM 12.0).

In the Tennessee River Embayment of Watts Bar Lake (downstream from the Clinch River), the volume of bottom sediments increased in each reach for the period of record of sediment range surveys, except in reaches near Watts Bar Dam (TRM 529.9 to 532.1) and about midway between Watts Bar Dam and the mouth of Clinch River (TRM 546.2-549.9). In the two cited reaches, sediment volume decreased between 1951 and 1956; the losses in the two reaches were more than offset by gains occurring in other reaches. At most sediment ranges in the Tennessee River Embayment of Watts Bar Lake, shifts in sediment volume (gains or losses) occurred in the deeper parts of the section.

High rates of increase in volume of bottom sediments in the Tennessee River Embayment of Watts Bar Lake occurred generally in the downstream portions of the reservoir, downstream from TRM 543.7 (Table 2). Moderate rates of increase have occurred in the reaches between TRM 549.9 and 557.1. In other reaches, the rate of increase in volume of bottom sediments has been relatively low.

In Chickamauga Lake sediment volume increased in all reaches between 1940 and 1947. In the next period between sediment range surveys, 1947-54, volume decreased in all reaches but one (4.1 to 7.2 miles upstream from Chickamauga Dam); the magnitude of losses was nearly the same or greater than the gains in the preceding period, especially in the upstream two-thirds of the reservoir (upstream from TRM 490.4). After 1954, the volume increased slightly in about three-fourths of the reaches in the reservoir. In the upstream half of the reservoir, changes in volume, either losses or gains, were appreciably less than changes in the downstream part throughout the period of record.

Channel alignment seems to influence the transverse distribution of bottom sediments in a section. Zones of greatest deposition in a section occur in that portion affected by eddies in the flow. Eddies are created as the 'live' flow separates from a curving channel boundary. The size of the eddy depends on the degree of curvature in the boundary and on the length of the curve. Bends, islands, and constrictions in a channel create eddy zones. Water in the eddy zone is in slow rotation with less velocity and turbulence than in the main stream of the flow. With the decrease in dynamic forces acting to suspend the sediment particles, more deposition takes place in the eddy zones than in the live stream of the river.

In the Clinch River, the influence of bends on the distribution of sediment deposits in a section has been observed in the TVA sediment ranges located at CRM 1.3, 4.7, 7.5, and 16.9. Islands in the channel also have affected deposition patterns in the Clinch River. The influence of the flow pattern on sediment deposition through one bend in the Clinch River is shown in Fig. 6. The bend is located between CRM 5.4 and 6.3, and is one of the sharpest bends in the study reach.

Deposition patterns in the Tennessee River Embayment of Watts Bar Lake are influenced by channel alignment, also. Eddy zones created by islands and constrictions in the channel affect transverse distribution of sediment at more than half the sediment ranges.

The influence of channel alignment on deposition patterns in Chickamauga Lake is not so apparent as in Watts Bar Lake. Apparently the curvature of the bends is so large that pronounced eddy zones are not created. The transverse slope of the channel bed seems to have more influence. Little or no gain in volume of bed material occurs in the steepest sloping portions of the bed.

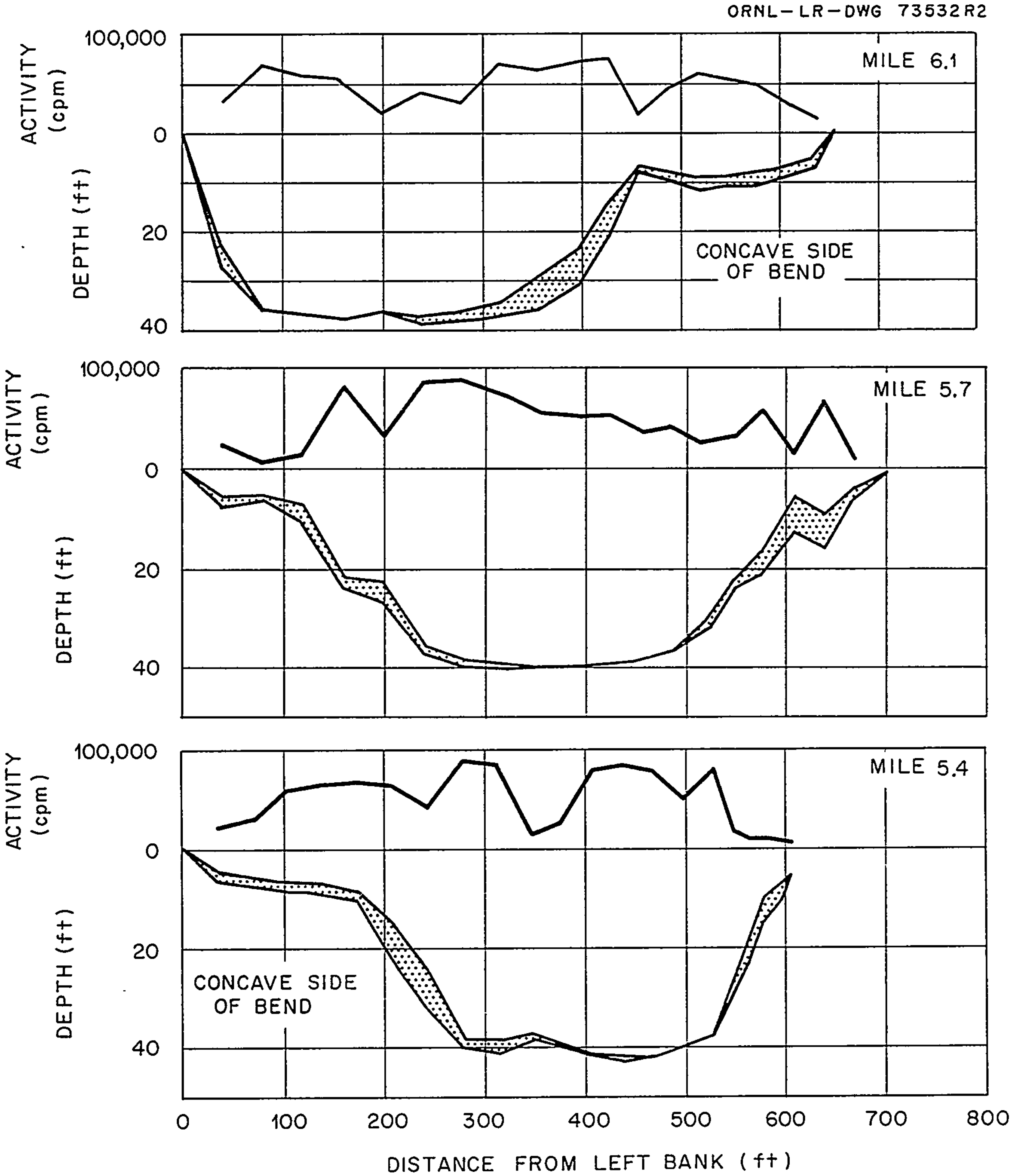

Fig. 6. Variation of Water Depth, Probed Depth of Bottom Sediment, and Gross Gamma Radioactivity Detected at Surface of Bottom Sediment in Cross Sections at CRM 5.4, 5.7, and 6.1

RELEASE OF RADIONUCLIDES FROM OAK RIDGE NATIONAL LABORATORY

Much of the liquid waste products at ORNL originate from various chemical processes used to extract a specific radionuclide, or group of radionuclides, from nuclear fuels removed from a reactor. In 1943 and 1944, the primary efforts were directed toward extracting plutonium from fuel irradiated in the graphite reactor. These extractions were made in the Chemical-separations Pilot Plant. In 1945 the RALA program for extraction of ^{140}Ba and ^{140}La began in the Hot Laboratory and Fission Products Separation Building. In 1947 a number of fission-product extraction processes, at pilot-plant scale, were initiated at ORNL. Many of these processes were in the Chemical-separations Pilot Plant. Later activities at the Laboratory diversified and other specialized facilities were constructed to house experiments using radioactive materials. A listing of major facilities producing radioactive waste at ORNL is given in Table 4.

The effect of changes in methods of disposing of liquid wastes released from facilities processing radionuclides may be discerned in the records of the release of radioactive waste waters from White Oak Lake (see Table 5). For instance, a decrease in the loads of all radionuclides between 1949 and 1950 resulted when treatment of waste waters in the Evaporator (Fig. 7) began. The loads of ^{137}Cs, ^{60}Co, and ^{144}Ce in 1955 and 1956 suddenly increased due to draining White Oak Lake in October 1955. The release of ^{144}Ce from the lake bed occurred in 1955 because either this particular radionuclide was not strongly sorbed in the sediments on the lake bed or because sediments containing this radionuclide were at the surface of the lake bed. The cesium and cobalt releases accompanied heavy runoff in the early part of 1956.

Until 1949, the waste-disposal treatment procedure at ORNL could be described as a triple settling process[14]: (1) storage in underground tanks permitting precipitation and decay of short-lived radionuclides, (2) deposition of precipitates in the Settling Basin, and (3) settling of solids in the Intermediate Pond and White Oak Lake

Table 4. Facilities Producing and Processing Significant Quantities of Radioactive Waste Materials at ORNL

Facility	Year Operation Began
Graphite Reactor	1943
Chemical-Separation Pilot Plant	1943
Chemical Laboratory	1943
Tank Farms	1943
Retention Pond (Equalization Basin locale)	1943
White Oak Lake	1943
Hot Laboratory and Fission Products Separation Building	1944
Settling Basin	1944
High Radiation Level Chemistry Laboratory	1948
Evaporator	1949
Metal Recovery Plant	1951
Radioisotopes Production Area	1951
Sewage Treatment Plant	1951
Seepage Pits	1952
Decontamination Laundry	1955
High Radiation Level Analytical Facility	1956
Process Waste Water Treatment Plant	1957
Equalization Basin (Process Waste System Basin)	1957
Fission Products Development Laboratory	1957

Table 5. Yearly Discharges of Radionuclides to Clinch River (curies)[16,a]

Year	Gross Beta	^{137}Cs	^{106}Ru	^{90}Sr	TRE(−Ce)	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co
1949	718	77	110	150	77	18	180	22	77	
1950	191	19	23	38	30		15	42	19	
1951	101	20	18	29	11		4.5	2.2	18	
1952	214	9.9	15	72	26	23	19	18	20	
1953	304	6.4	26	130	110	6.7	7.6	3.6	2.1	
1954	384	22	11	140	160	24	14	9.2	3.5	
1955	437	63	31	93	150	85	5.2	5.7	7.0	6.6
1956	582	170	29	100	140	59	12	15	3.5	46
1957	397	89	60	83	110	13	23	7.1	1.2	4.8
1958	544	55	42	150	240	30	6.0	6.0	8.2	8.7
1959	937	76	520	60	94	48	27	30	0.5	77
1960	2190	31	1900	28	48	27	38	45	5.3	72
1961	2230	15	2000	22	24	4.2	20	70	3.7	31
1962	1440	5.6	1400	9.4	11	1.2	2.2	7.7	0.36	14
1963	470	3.5	430	7.8	9.4	1.5	0.34	0.71	0.44	14

[a]Values calculated from data supplied by Applied Health Physics Section, ORNL.

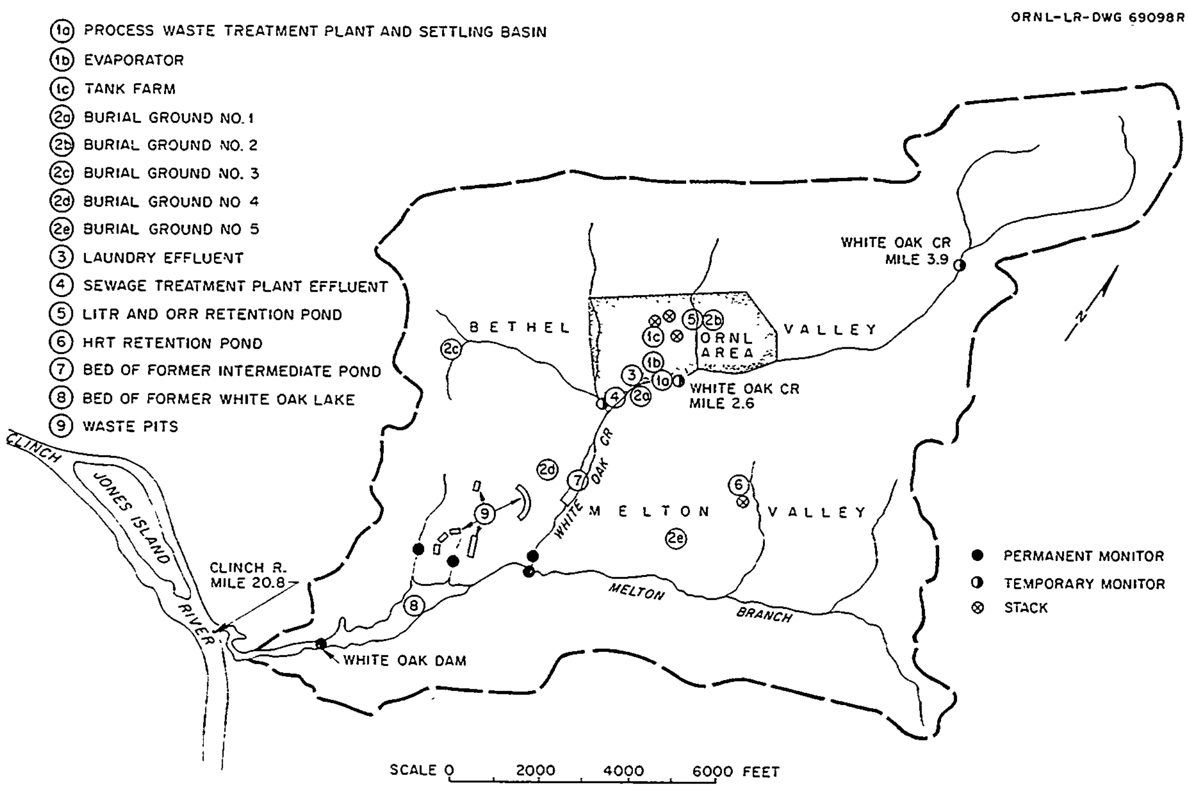

Fig. 7. Map of White Oak Creek Basin Showing Sources of Radioactive Contamination and Location of Stream Monitoring Stations

(flood destroyed Pond in 1944). In addition to triple settling supernatant from the tanks mixed with process-waste waters for dilution within the plant area at ORNL and effluents from the Settling Basin mixed with waters of White Oak Creek and the Clinch River for dilution outside the plant area. "This method..of waste disposal..was entirely satisfactory because the dilution afforded by the Clinch River lowers the (radioactive) liquid waste concentration to acceptable levels"[15].

In 1949, the Evaporator, the first major modification to waste treatment, was placed in operation. Supernatant from the storage tanks fed into the Evaporator and its condensates flowed into White Oak Creek. The Evaporator was an interim process in waste treatment, being decommissioned in 1954. In 1952, a pit dug in Conasauga Shale residuum demonstrated that radionuclides could be removed from waters seeping through the banks and walls of the pit by filtration and sorption in the soil. These seepage pits (trenches) constituted the method of waste disposal which soon supplanted the Evaporator. Seven pits have been put in operation since 1952.

In 1955, the accumulation of radionuclides in White Oak Lake presented problems because: (1) the lake had become a habitat for migratory birds and fish, and (2) radionuclides in lake waters had come to equilibrium with radionuclides deposited or sorbed on lake bed[15]. For these reasons the lake was drained in October 1955. The drained lake provided a facility for emergency storage in the event of an unusually high-level release of radioactivity.

Growth and diversification of activities at ORNL created many sources of release of low-levels of radioactivity to the waste water system. To control this problem the Process Waste-Water Treatment Plant was constructed in 1957.

The release of ^{106}Ru from White Oak Lake had been almost negligible prior to 1959. As a result of leakage from the seepage pits subsequent to the transfer of a large quantity of this radionuclide to the pits, the release increased very substantially in 1959, 1960, and 1961.

There are many other fluctuations in the loads of radionuclides released from White Oak Creek that cannot be easily documented. The inability to document the fluctuations is due to the complexity of operations at ORNL.

ANNUAL MONITORING SURVEYS OF RADIOACTIVITY IN BOTTOM SEDIMENTS

Annual monitoring surveys have been made since 1951 at selected cross sections between Clinch River Miles 1.1 and 27.5 and between Tennessee River Miles 354.5 and 570.8 (note locations of tributaries and dams in figures 1 and 2 and in table 1 and locations of the observation sections in figures 1, 3, 4, and 8). In 1952 and 1961 the surveys extended downstream on the Tennessee River to TRM 24.6; in 1961 the survey was extended upstream on the Clinch River to CRM 39.1. In each survey the flounder, a Geiger-Mueller tube detector, was used to measure radiation levels at the surface of the sediment at about ten equally-spaced points in each section. Beginning in 1954 the survey included sampling of the upper portion of the sediment at flounder observation points with an Ekman dredge. Sampling at every point was not always possible because of encountering bedrock or large-sized particles (gravel or cobbles). The point samples for each section were composited and radiochemical analyses were made (see Tables 8-19 for results of radiochemical analyses, Appendix).

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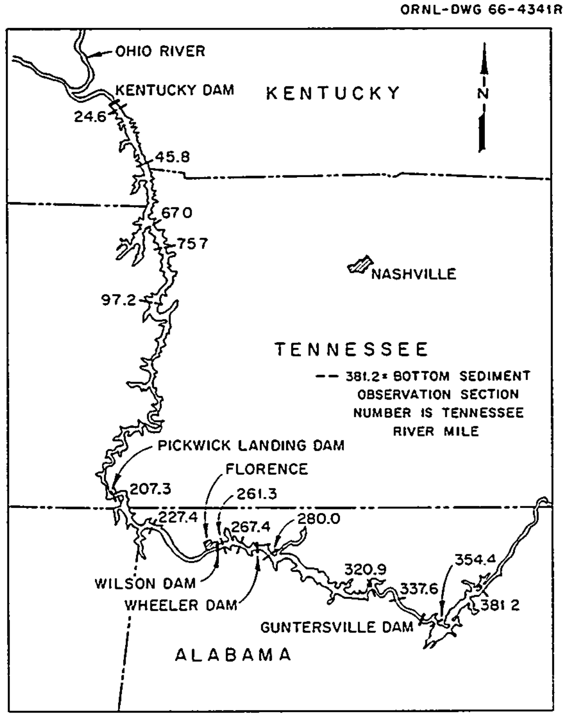

Fig. 8. Map of Tennessee River Showing Bottom Sediment Observation Sections in Kentucky, Pickwick Landing, Wilson, Wheeler, and Guntersville Lakes

Before each survey the flounder was calibrated for attenuation of cosmic radiation by water shielding, and dredge samples from Fort Loudoun Lake provided sediment for an estimate of background concentrations of specific radionuclides.

Get there

Descriptions of instrumentation and of methods of conducting surveys and of analyses of results of the surveys are given in greater detail in reports by Garner and Kochtitzky[5] and by Cottrell[6].

Longitudinal Distribution

Flounder data from the 1958 and 1961 surveys illustrate general patterns of distribution of radioactivity in bottom sediment of the Clinch and Tennessee Rivers (Figs. 9 and 10). Flounder count rates observed in 1958 were generally higher than rates for preceding or subsequent surveys; data from the 1961 survey are the most recent available.

Highest average count rates in the Clinch River have been confined to the reach extending from CRM 5 to 15. The annual maximum

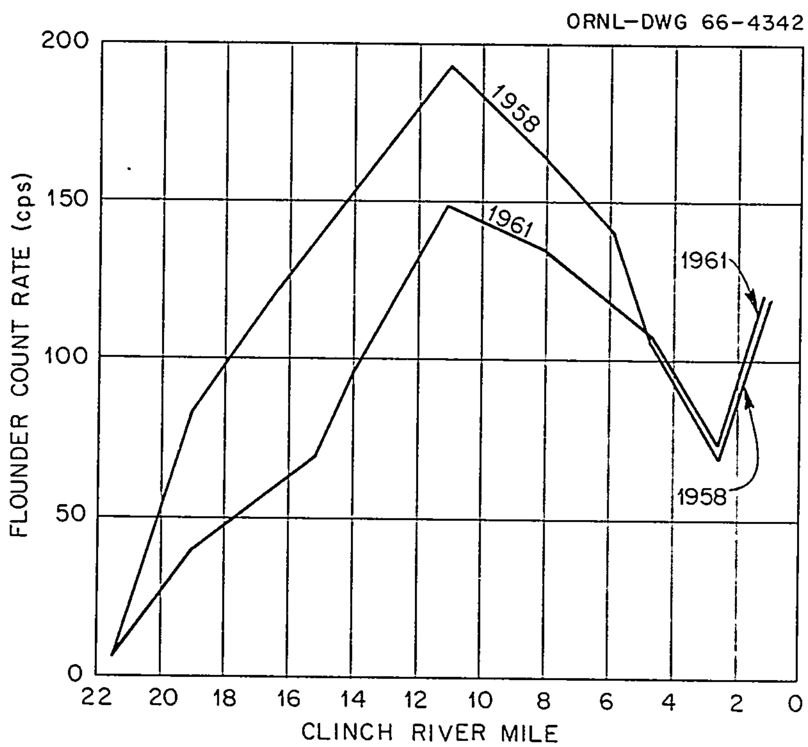

Fig. 9. Variation of Average Flounder Count Rate in Clinch River, Surveys of 1958 and 1961.

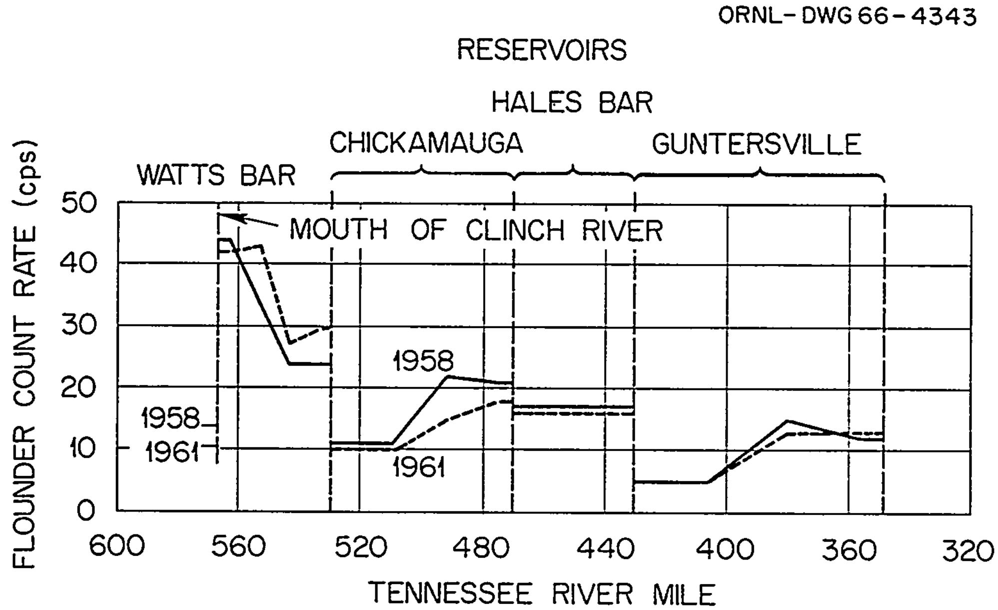

Fig. 10. Variation of Average Flounder Count Rate in Tennessee River, Surveys of 1958 and 1961.

average count rate shifted somewhat from year to year, generally occurring at either CRM 8.0 or 10.0. The centroid of the longitudinal distribution of radioactivity between CRM 5 and 15 does not appear to shift appreciably as the radionuclide loads released from White Oak Lake change from year to year.

In most years average flounder count rates upstream from CRM 14 or 15 were lower than those in reaches downstream. Accumulation of sediment deposits in this portion of the river has been less than in the portion of the study reach downstream from CRM 14 or 15. Lack of accumulation of sediment deposits, especially finely divided particles, upstream from these sections is the probable reason for observing lower levels of radioactivity (see section on Physicochemical Characteristics).

Low count rates have been observed at CRM 2.6, relative to those for sections at CRM 1.1 and 4.7, in each annual flounder survey. The decrease in average count rate between CRM 4.7 and 2.6 is greater than that to be expected from volumetric dilution due to inflow from the Emory River (24 percent of flow in Clinch River). Count-rate measurements made at CRM 2.6 may not be representative of the general longitudinal distribution of radioactive sediments in this region of the river. The measuring section is immediately downstream from the cooling-water outlet from Kingston Power Plant, and is also immediately downstream from a small tributary entering from the right bank. At a section located immediately upstream from the cooling-water outlet, radiation levels are higher than at the 'standard' measuring section for CRM 2.6 and are about 16 percent less than those observed at CRM 4.7.

Hemphill and others compared various methods of determining longitudinal distribution of radioactivity in the Clinch River (Fig. 11)[2]. Data used in comparison included average flounder count rate at the surface of the sediment, average gross gamma count of one-inch segments of cores, and radionuclide content of composites of the cores. Later, R. W. Andrew[17] superposed results of radiochemical analyses of total identified radioactivity in dredge samples (collected by USPHS

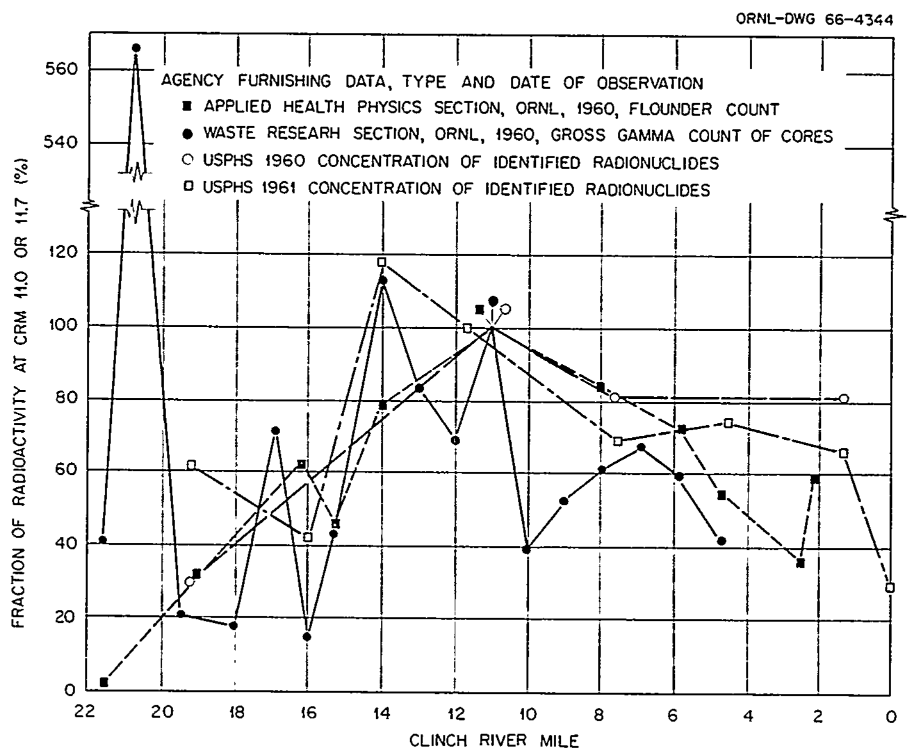

Fig. 11. Comparison of Methods for Determining Longitudinal Distribution of Radioactivity

personnel) on the comparative plot, Fig. 11. The patterns in Fig. 11 are much alike for all methods. Differences in patterns result primarily from differences in location of sampling sections. Only one survey in 1960 included observations at CRM 20.8.

Radionuclide concentration in bottom sediment of the Tennessee River appears to decrease exponentially for standardized concentration of ^{137}Cs (Fig. 12). Standardized concentrations are estimates of the concentration for sections located 3 miles upstream from the dams.

Garner and Kochtitzky[5] concluded that in the 1952 survey the flounder was detecting only variations in the radioactivity of naturally occurring radionuclides in bottom sediment downstream from Hales Bar Dam (TRM 431.1). Therefore, variation in concentration of ^{137}Cs rather than that of flounder count rate is used to study longitudinal distribution of radioactivity in the Tennessee River (Fig. 13).

There is a tendency in the Tennessee River for radioactivity to increase in concentration in the downstream direction within a reservoir (see Fig. 13). Cottrell[6] reasoned that the increase was the result of scouring action in the first 20 miles of reach downstream from the dam and deposition farther downstream into the reservoir.

The general pattern of longitudinal distribution of radioactivity in the Tennessee River, shown in Fig. 13, has remained about the same from 1954 through 1961 in each reservoir upstream from TRM 354.5.

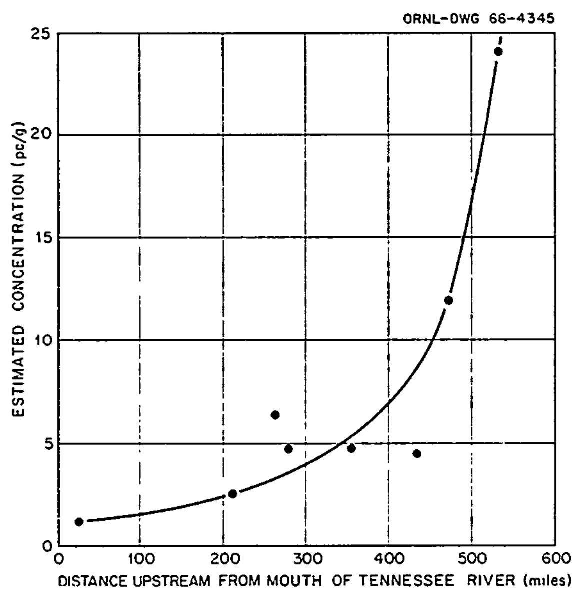

Fig. 12. Variation in ^{137}Cs Content, Tennessee River Bottom Sediment in 1961

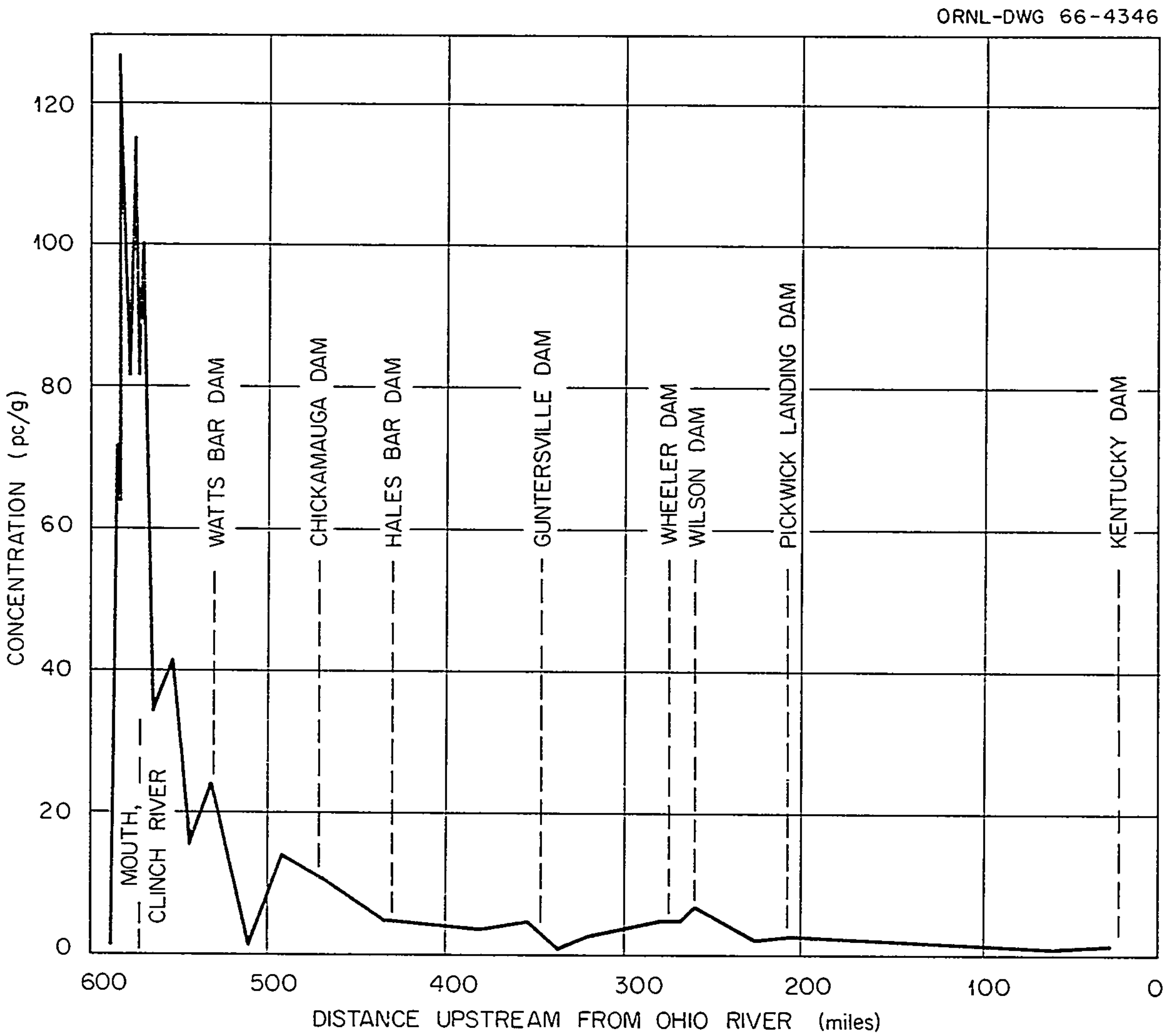

Fig. 13. Longitudinal Distribution of ^{137}Cs in Clinch and Tennessee Rivers in 1961

Influence of Dispersion and Flow Pattern

Releases from White Oak Creek remain close to the right bank of the Clinch River for more than a mile downstream. Lateral mixing of waters from White Oak Creek is complete at distances somewhere between 4 and 6 miles downstream from the mouth of the creek[1, 3, 18]. Parker investigated factors influencing development of complete lateral mixing, particularly temperature differences between waters of the creek and river, in 1957 and in 1959[18, 19]. Through a series of tracer tests he found that waters from White Oak Creek generally passed through the right channel of the river at Jones Island (Fig. 1) and remained in the right portion of the channel for a considerable distance downstream from the island.

Incomplete lateral mixing of waters from White Oak Creek has an effect on the dispersion of radioactive materials in bottom sediment in the uppermost reaches of the study area. At CRM 19.4 and 20.7 the highest flounder count rates occur in the right third of the channel bed (see Fig. 10, Ref. 6).

Farther downstream in the Clinch River distribution of radioactivity across the section seems to be influenced by the local flow pattern. In particular eddy zones in and about bends, islands, and submerged ridges affect the distribution. Count rates are higher in the part of the stream bed which is in or near the convex side bends (CRM 4.7, 5.8, 6.9, 9.4, 12.5, 14.0, and 15.2, Fig. 1). Concentration of radioactivity in the sediment is fairly uniform across the sections, or is slightly higher at mid-section, at CRM 8.0, 11.0, and 16.3. There are small slightly exposed islands or slightly submerged ridges in the vicinity of these sections.

In the Tennessee River distribution of radioactivity in bottom sediments across sections is more uniform than for sections in the Clinch River. Nevertheless channel alignment has an effect on the distribution pattern, but the effect is less marked than in the Clinch River (see Figs. 3, and 10-14, Ref. 6).

Limits of Detection

Presumably bottom sediments in the Clinch River upstream from the mouth of White Oak Creek should not be contaminated by fission-product releases from White Oak Lake. Sediments at CRM 27.5 do seem to be free of contamination by these releases. At CRM 21.5, 0.7 mile upstream from the mouth of White Oak Creek, unusually high flounder count rates were obtained in surveys of 1957, -58, and -59. Contamination of bottom sediments with fission-product releases apparently occurred in the river upstream from the creek, at least as far as CRM 21.5. During short periods of essentially no flow, contaminated material could have been carried in waters of the Clinch River upstream from White Oak Creek either by adverse velocity gradients or by wind action. In 1961, radioactive contamination of the river bed sediment due to releases from White Oak Creek did not extend upstream to CRM 24.1, nor was there pronounced contamination at CRM 21.5.

From study of available data it is concluded that the bottom sediments of the Tennessee River upstream from the mouth of the Clinch River at TRM 570.8 (Fig. 3) have not been contaminated by fission-product releases during the period 1954-61.

Samples of bottom sediments collected in the Clinch River at CRM 21.5, in the Tennessee River at TRM 570.8, and in Fort Loudoun Lake in 1961 are assumed to be uncontaminated by releases from White Oak Lake. The average concentration of each radionuclide in samples from these sections is considered to represent the background concentration for the radionuclide. Concentrations of ^{106}Ru in composite samples obtained in the annual monitoring survey of 1961 (Tables 16, 17, Appendix) were greater than background (2.4 pc/g) in the reach extending from the mouth of White Oak Creek on the Clinch River to near the mouth of the Tennessee River, near Paducah, Kentucky.

Concentrations of ^{137}Cs, ^{90}Sr, ^{144}Ce, and ^{60}Co were found to be greater than background (respectively 1.3, 0.18, 0.73, and 0.34 pc/g) in Pickwick Landing Lake. Concentrations of ^{95}Zr and ^{95}Nb, trivalent rare earths, and ^{90}Y were higher than background upstream from Guntersville Dam (backgrounds, respectively, were 0.73, 1.0, and 1.0 pc/g).

As Garner and Kochtitzky[5] have pointed out, the concentrations of fission products in the bottom sediments are not enough greater than concentrations of naturally occurring radionuclides in sediments to cause a significant response in the detector of the flounder. Very low-level contamination detected by the founder is almost exclusively that due to gamma emissions of ^{137}Cs. The concentration of ^{137}Cs must be greater than 4 pc/g in order to cause a significant response in flounder count rate. Concentrations of ^{137}Cs have been generally less than 4 pc/g in bottom sediments downstream from Guntersville Dam (see Table 9, Appendix).

RADIONUCLIDE CONTENT OF UPPER PORTION OF BOTTOM SEDIMENTS

Relationship of Concentration in Sediment to Annual Load Released from White Oak Lake

Annual variations in concentrations of radionuclides in the upper portion of bottom sediments appear to follow the same patterns of variations as annual releases of these radionuclides from White Oak Lake. For example, the concentration of ^{137}Cs in composite samples of bottom sediment from CRM 1.1 (from annual monitoring surveys) varies almost directly with the total quantity of this radionuclide released from White Oak Lake during the preceding year (July 1 to June 30) for the period 1954-61 (see Fig. 14)[4]. The correlation index is 0.80. Correlation indices for other principal radionuclides, found in bottom sediment at this section of the Clinch River, are listed in Table 6. Annual changes in the concentration of most radio-

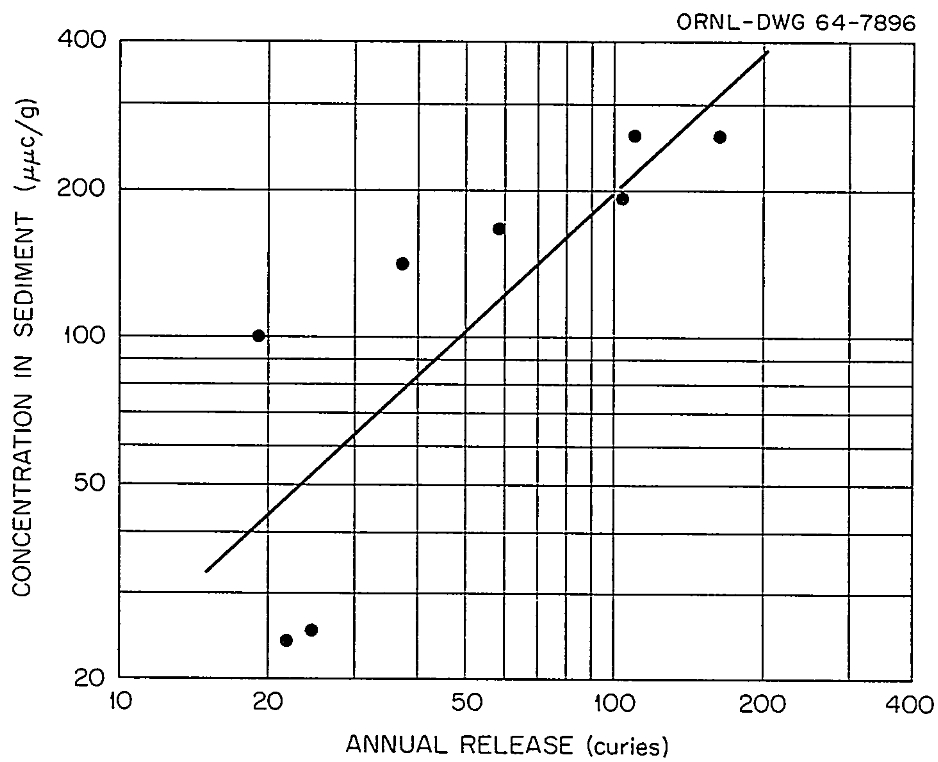

Fig. 14. Relationship of Annual Changes in ^{137}Cs Concentration in Bottom Sediments at CRM 1.1 to Changes in Annual Load of ^{137}Cs Released from White Oak Lake in Year Beginning July 1 and ending June 30

Table 6. Correlation Indices for Comparison of Annual Determinations of Radionuclide Concentrations in Bottom Sediment at Selected Sections in the Clinch and Tennessee Rivers to Annual Loads Released[a]

Location (CRM)	Correlation Index					
	Radionuclide					
	^{137}Cs	^{106}Ru	^{144}Ce	^{90}Sr	^{60}Co	Rare Earths[b]
19.1	0.87	0.76[c]	0.41[d]	0.62[c,d]	0.51[c,d]	0.69
16.3	0.96	0.97	0.40[d]	0.81	0.62[c]	0.89
15.2	0.90	0.94[c]	0.75[c]	0.92[c]	0.79	0.90
14.0	0.90	0.95	0.57[d]	0.79	0.48[d]	0.92
11.0	0.93	0.98	0.80	0.90	0.84	0.92
1.1	0.80	0.89	0.66	0.72	[e]	0.48[d]
(TRM)						
562.7	0.96	0.99	0.97	0.27[d]	0.70	0.89
432.0	0.92[c]	0.93	0.91	0.22[d]	0.78	0.47[d]
491.9	0.91	0.97[c]	0.73[c]	−0.11[c,d]	0.85[c]	0.66[c]

[a]Correlation of logarithms of radionuclide concentrations which were determined for annual monitoring surveys in period 1954–61. From records furnished by ORNL Applied Health Physics Section.

[b]Radiochemical analyses included ^{90}Y but exculded ^{144}Ce.

[c]No data for 1955 survey.

[d]Correlation index not significantly different from zero.

[e]Insufficient data for analysis.

nuclides in the upper portion of bottom sediments appear related to load of the radionuclides released annually through White Oak Dam throughout the study reaches in the Clinch and Tennessee Rivers (Table 6).

Two models describe possible means of incorporation of radionuclides in the bottom sediment in response to annual releases through White Oak Dam: (1) deposition during preceding 12 months of suspended sediments which sorbed radionuclides prior to their release through White Oak Dam (2) Or, sorption *in situ* by bottom sediments in the river of radionuclides released as dissolved ions[4].

Both models require either regular, continuous deposition of sediment over much of the sampled section, or release of fairly constant quantities of the radionuclides during the preceding 12 months. Model (2) requires rapid establishment of chemical equilibrium between water and sediment followed by each incremental layer of sediment being covered and sealed by other sediment. Quantities of radionuclides released from White Oak Lake have not been constant. Furthermore, deposition of sediment at sampling sections is not continuous across the entire width of the section (see Tables 2 and 3, Fig. 5). However, data for TVA sediment ranges show wide zones of continuous, net accumulation of sediment on the sides of the stream channel, and only areas along the thalweg of the stream channel are subject to scour, particularly in periods of high streamflow[4].

For releases of ^{137}Cs, more than 80 percent of the radionuclide passing through White Oak Dam is associated with suspended sediment[20]. The reaction by which cesium is sorbed on the sediment is not readily reversible[4]. Hence, incorporation of ^{137}Cs into bottom sediments can be assumed to conform to model (1).

Relationships in the Longitudinal Distribution of Radionuclides

Similarities in longitudinal distribution of certain radionuclides in the bottom sediments in 1961 annual monitoring survey appear to persist from the mouth of White Oak Creek on the Clinch River to the mouth of the Tennessee River (see Tables 8-19, Appendix). Correlation analyses have been used to study the relationships in longitudinal distribution of these radionuclides. In the correlation analyses the index of correlation, standard error of the estimate, and regression coefficient were computed for the comparison of logarithms of concentrations for all possible pairings of ^{137}Cs, ^{106}Ru, ^{60}Co, ^{144}Ce, ^{95}Zr, ^{95}Nb, ^{90}Sr, and trivalent rare earths (including ^{90}Y but excluding ^{144}Ce)[4]. In Fig. 15 the regression curves for the comparison of concentration of the indicated radionuclide to the concentration of ^{137}Cs are plotted, and the indices of correlation and standard errors of the estimate are listed.

The similarity in slopes for the regression curves in Fig. 15 between concentrations of ^{106}Ru, ^{60}Co, rare earths, and ^{137}Cs suggests that similar mechanisms control the longitudinal distribution of these particular radionuclides.

The concentrations of radionuclides in bottom sediments

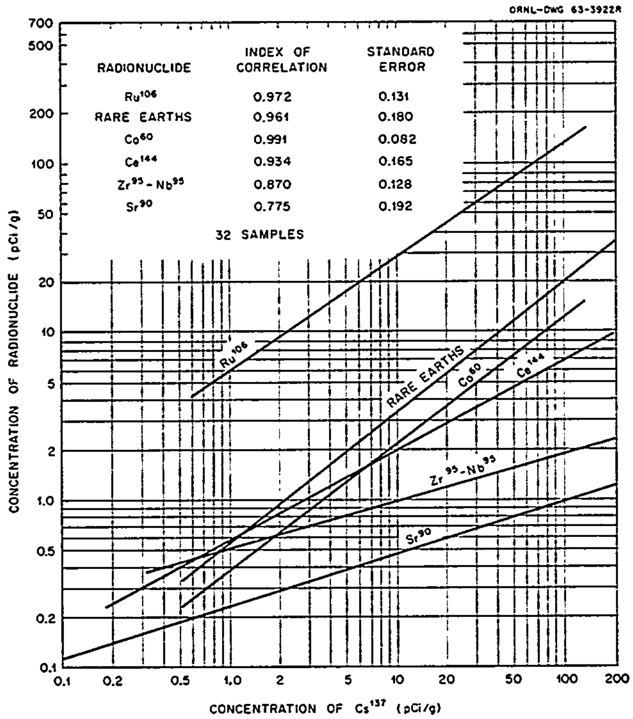

Fig. 15. Correlations Showing Similarity in the Longitudinal Distribution of Radionuclides in the Bottom Sediments of the Clinch and Tennessee Rivers

decrease in the downstream direction in the Tennessee River (see Fig. 16). The possibility exists that flow dilution is the cause for the decrease in concentration. In Fig. 16, the decrease in concentration to be expected due to flow dilution (discharge records for the period 1960-60 furnished by TVA and USGS) with distance downstream from Watts Bar Dam is shown by the line labelled "discharge". Other curves in the figure depict the relative change in concentration of a radionuclide with distance downstream from Watts Bar Dam (referred to its concentration for the first section upstream from Watts Bar Dam). Another possibility exists — that the decrease in radionuclide concentrations in the downstream direction results from dilution of contaminated sediments by uncontaminated sediments (lowermost curve in Fig. 16). The expected dilution of contaminated sediments by uncontaminated sediments was computed from data on the fraction of the sediment in a reservoir that originated from the next upstream reservoir[5].

Based on curves in Fig. 16, the reduction in concentration of each radionuclide, except for ^{90}Sr, appears to be greater than

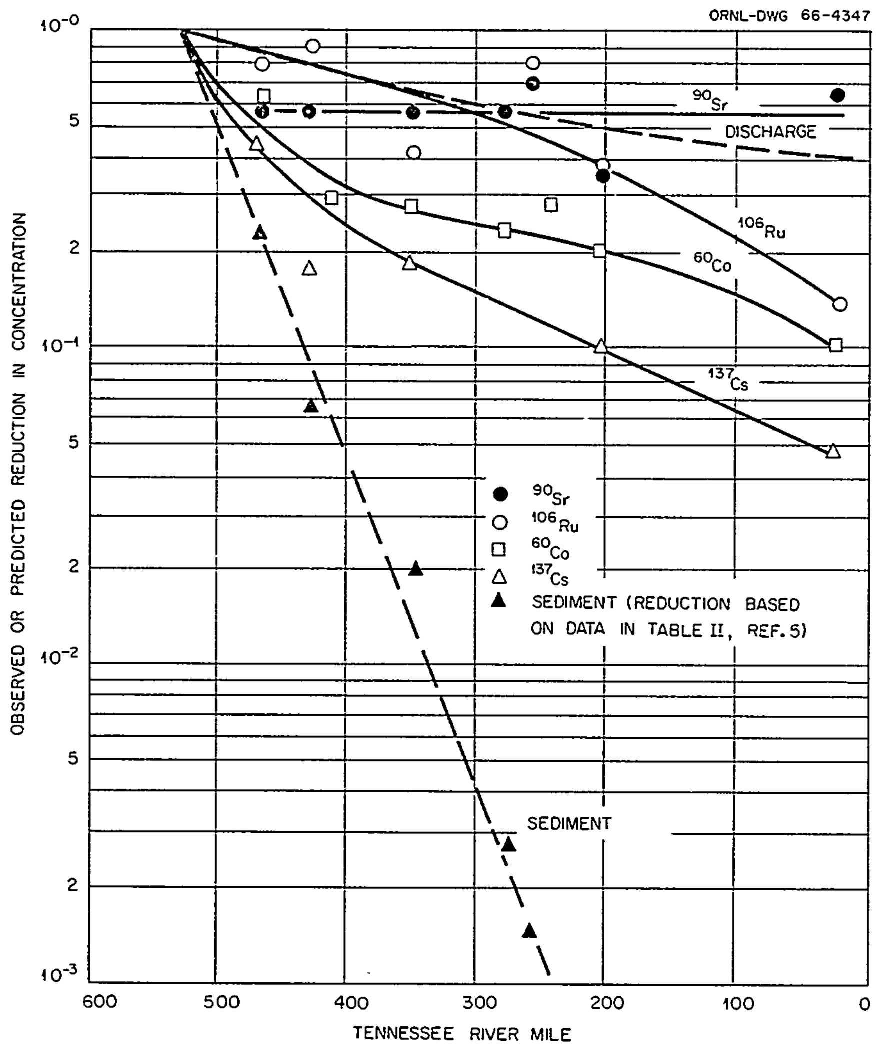

Fig. 16. Observed Reduction in Concentration of Radionuclides and Reduction Predicted by Flow or Uncontaminated Sediment Dilution

simple volumetric dilution but not as great as dilution by uncontaminated sediment.

Results of analysis in Figures 15 and 16 further suggest that the mechanism controlling the distribution pattern for several radionuclides is sedimentation rather than the chemical process of equilibration (reversible sorption) between radionuclide concentrations in the sediments and in the water. Furthermore, once these radionuclides are incorporated into bottom sediments the potential for their release through desorption is almost negligible[4].

Slopes of the curves for ^{95}Zr and ^{95}Nb and for ^{90}Sr are different from those for other radionuclides in Fig. 15. The differences in slopes of the curves imply that different mechanisms may be controlling the distribution of these three radionuclides in the bottom sediments. Longitudinal distribution of ^{90}Sr in the bottom sediments is quite similar to the distribution that would have resulted from the effects of flow dilution (Fig. 16). It is quite probable that the most important mechanism controlling the longitudinal distribution of ^{90}Sr is flow dilution (see section on Physicochemical Characteristics and Ref. 4).

Seasonal Changes in Distribution of Radioactivity

In general, levels of radiation in bottom sediments of the Clinch River tend to increase as the distance downstream from White Oak Creek increases (see Fig. 17). This tendency has been ascribed to hydraulic factors that may influence sediment deposition: larger flow areas, lower velocities, and greater surface area. However, the change in radiation levels with distance is not regular. In particular relatively high levels of radiation have been observed, in summer surveys, at several sections downstream from CRM 12[4].

Possibly the higher zones of radiation levels downstream from CRM 12 result from seasonal variations in these and other hydraulic factors; in the winter season greater discharges and increased velocities of flow occur; in the summer thermal stratification occurs when flows are less than 10,000 cfs (see Fig. 13, Ref. 4).

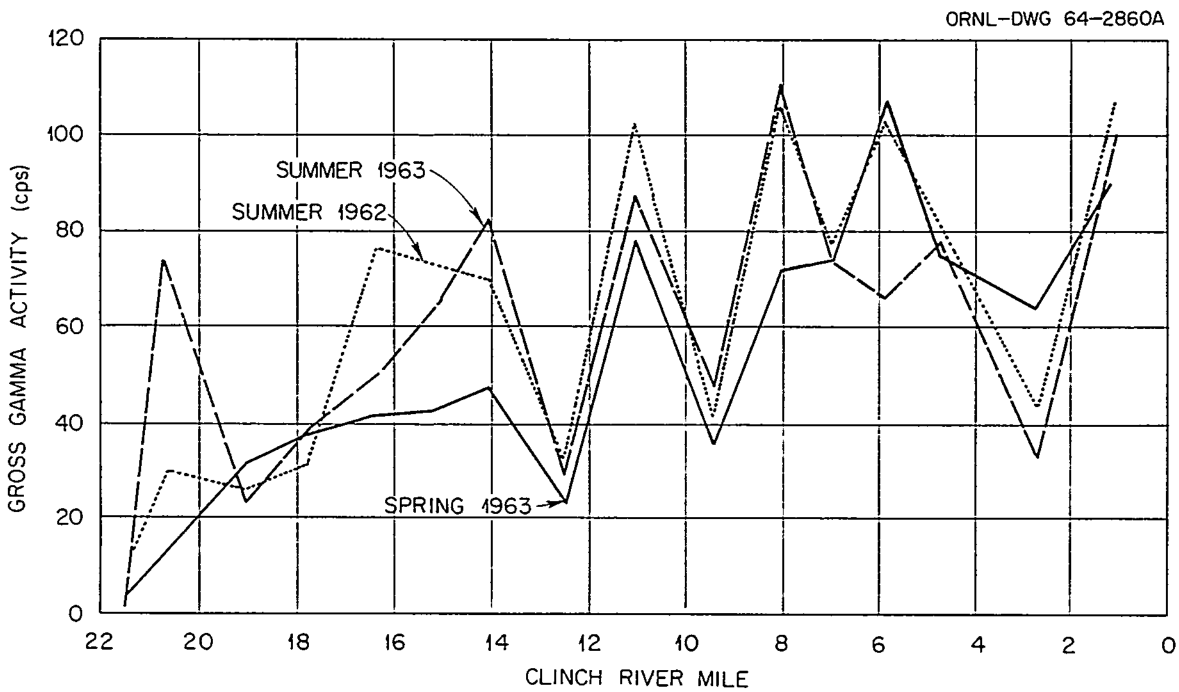

Fig. 17. Longitudinal Variation in Average Sectional Gamma Count Rate at the Surface of Bottom Sediments in Clinch River

A survey of gamma radiation at the surface of bottom sediment, by ORNL Waste Disposal Research Section and USGS, in the spring of 1963 (April 30 to May 8) explored the possible influence of the seasonal changes in the various hydraulic factors on distribution of radioactive sediments. This spring survey (1963) was made as soon after the winter high flows as practicable using the method reported by Cottrell[6].

The results of three gamma radiation surveys shown in Fig. 17, summer surveys of 1962 and 1963 and spring survey of 1963, seem to indicate that the decrease in effective flow area due to thermal stratification in the summer period does not cause any major change in the longitudinal variation of radioactivity in the bottom sediments[4].

Vertical Distribution of Radioactivity in Upper Strata of Bottom Sediments

Limited information is available on the vertical distribution of radioactivity in bottom sediments of the Clinch River from gross gamma counting of cores, collected in the summer of 1960 (see Description of Available Data). Maximum depth of penetration of the coring tool used for the sampling was 14 inches. In addition to the short depth of penetration available, plugging of the core barrel frequently occurred. Hence, information on the vertical distribution of radioactivity was restricted to a relatively thin layer of the uppermost strate of bottom sediments (average length of sample 7-1/2 inches).

Some variation in radioactivity with depth was noted in practically all of the 107 cores collected from the nineteen sections, CRM 4.7 to 22.5, sampled in 1960. The distribution of radioactivity in some cores seemed to be related to increases and decreases in the annual loads of radionuclides released through White Oak Dam. For example, similar patterns of vertical distribution in gross gamma count occurred in three cores collected at CRM 9.0 (Fig. 18). These patterns were similar to the pattern of gross gamma radioactivity released annually through White Oak Dam (inset, lower left corner,

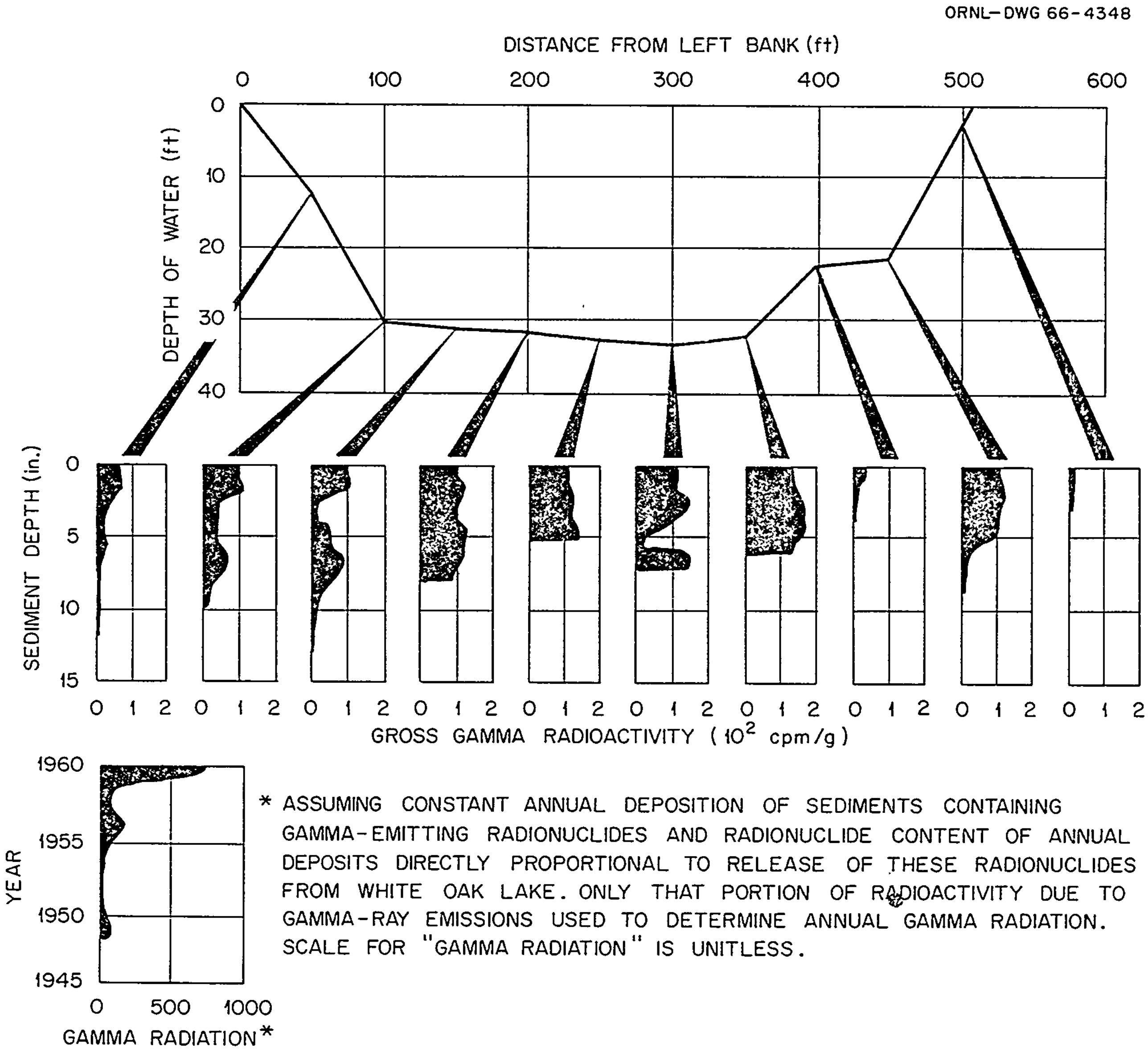

Fig. 18. Vertical Distribution of Radioactivity at Indicated Verticals and Cross-Sectional Shape at CRM 9.0, and Simulated Vertical Distribution of Radioactivity for Constant Deposition of of Releases from White Oak Lake

Fig. 18).

A study of all plots of vertical distribution of radioactivity in cores, showed that radioactivity levels neared background levels where the water depth over the coring hole, at the time of coring, was less than five feet, in sections downstream from CRM 16.9. From November to April in each year water levels in the river are nominally five to six feet lower than the levels at the time of coring (summer 1960). Low radioactivity levels in shallow-water portions of the sampling section may result from shorter duration of contact with radioactive waters of the Clinch ('summer' period) River and, also, may result from subaerial erosion of exposed sediments in the 'winter' period.

Effects of Flow Area and Channel Curvature on Distribution of Radionuclides

R. W. Andrew[21] was first to define a relation between flow area and the concentration of radionuclides in bottom sediments for the Clinch and Tennessee Rivers (Fig. 19). Andrew described the

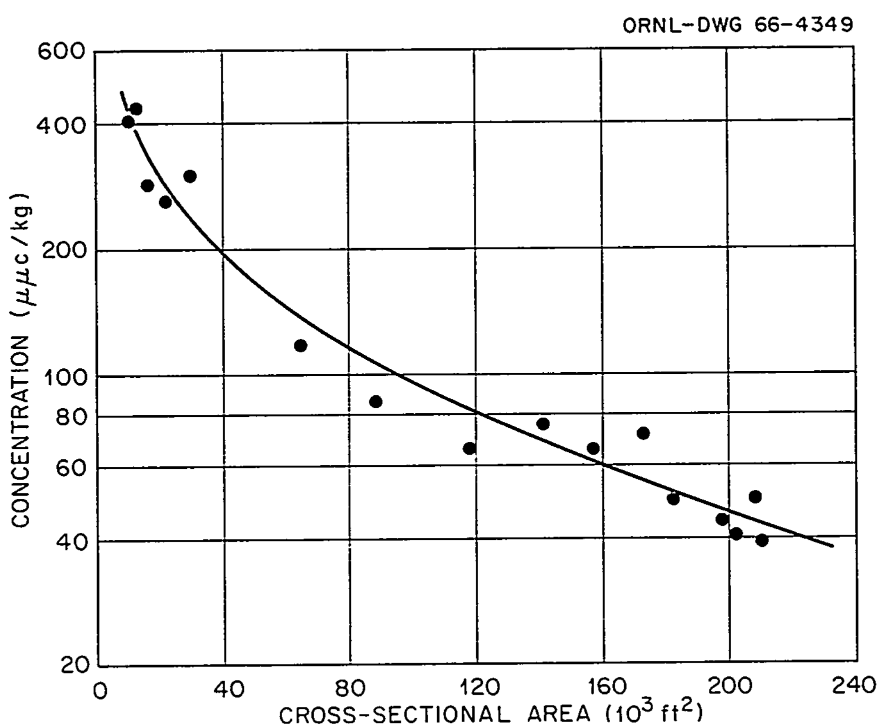

Fig. 19. Variation of Total Identified Radionuclide Concentration with Flow Area

relation in Fig. 19 as defining a dilution effect. He, as Cottrell[6], believed that if the radioactivity is dispersed over a larger area (surface area of the bottom sediments) the concentration of the radioactivity must be less.

The flow area, used to develop the relationship in Fig. 19, is normal to the channel-bed surface area. Hence, the flow area is not a direct expression of either 'dilution' or surface area. However, geometrical properties of sections in the Clinch and Tennessee Rivers are such that the length of perimeter for the sections generally increases as the flow area of these sections increases. The product of perimeter length and longitudinal distance is the surface area of the channel bed. Thus, flow area for the study reaches in the two rivers is an indirect measure of surface area.

In previous sections of this report the influence of channel geometry, particularly curvature, on the deposition of sediment and on the distribution of radioactivity in these sediments has been discussed. R. W. Andrew[21] delineated a possible relationship between radionuclide content of the bottom sediments and channel curvature. He found that the concentration of radionuclides in the upper portion of bottom sediments increased as the radius of curvature increased. A definitive relationship between concentration and curvature was not established because information was lacking on effects of other factors (particle size, position of the sampling section relative to beginning of bend).

INVENTORY OF RADIONUCLIDES IN SEDIMENTS

Complete inventory of radioactivity in the bottom sediments of the Clinch and Tennessee Rivers has not been made prior to work reported in Part B of this report. The quantity of radioactive materials associated with these sediments might be ascertained by utilizing data on radionuclide concentrations obtained in annual monitoring surveys and on sediment deposition or by radiochemical analyses of cores collected in these river sediments[4].

Inventory of Radionuclides in Upper Portion of Clinch River Bottom Sediments

Based on estimates of radioactivity in the bottom sediments of the Clinch River, from results of core sampling program in 1960, the total quantity of radioactive material accumulated is small relative to the quantity of radioactivity released through White Oak Dam.

The following quantities of radionuclides, in curies, were found to be associated with the upper 6-10 inches of sediments: 43.2 for ^{137}Cs, 14.7 for rare earths, 13.2 for ^{106}Ru, 4.7 for ^{60}Co, and 0.7 for ^{90}Sr; the total is 76.5 curies. This inventory was made for the reach extending from CRM 4.7 to 21.5 (essentially between the mouths of Emory River and White Oak Creek). Computational methods of obtaining this inventory and more detailed results of the work were reported by Morton[2].

Through review of sediment range data furnished by TVA the average sediment thickness is estimated to be three to four times greater than that sampled by the coring tool used in the 1960 inventory work. For this reason it might be assumed that the total quantity of radioactivity in the bottom sediments is a few times greater than the 76.5 curies determined as a result of the coring done in the summer of 1960.

The inventory could be on the order of 200 curies. A comparison with the quantities of radionuclides released through White Oak Dam, listed in Table 5, will show that the relative retention of these radionuclides in the bottom sediments is small.

PHYSICOCHEMICAL CHARACTERISTICS OF BOTTOM SEDIMENTS

Particle-Size Distribution in Bottom Sediments

Two separate determinations were made by the U. S. Geological Survey of the particle-size distribution in cross-sectional samples composited from bottom sediment cores collected during 1961 at five sections in the Clinch River. The averages of the results of those determinations (Table 20, Appendix) indicate the bottom sediment in the Clinch River to be a well graded silty loam with a rather constant content of clay-size material. The minimum median particle size occurred at CRM 11.9.

The quantity of radionuclides sorbed on river sediment should increase with decreasing particle size of the sediment for two reasons: (1) because the relative content of clay minerals, the mineral group with the highest sorption capacities for many cations, usually increases as the median particle size of the sediment decreases; (2) because in many minerals, including the clay minerals, the exchange capacity varies directly with the surface area, which increases as particle size decreases.

Relationship between particle-size distribution and radionuclide concentrations (ORNL radiochemical analysis) in the sediment with distance for the five river cross sections sampled is shown in Fig. 20. Radionuclide concentrations vary more widely from section to section than does particle-size distribution and do not appear to bear the direct relationship to particle-size distribution that might be expected on the basis of the statement made in the preceding paragraph.

Comparison plots of particle-size distribution and ^{137}Cs content of bottom sediment samples collected in a single survey of the Clinch and Tennessee Rivers by the USPHS have been made by R. W. Andrew and are shown in Fig. 21[22]. A possible correlation exists between the percent clay-size particles in the bottom sediments and the cesium content of the samples studied by Andrew.

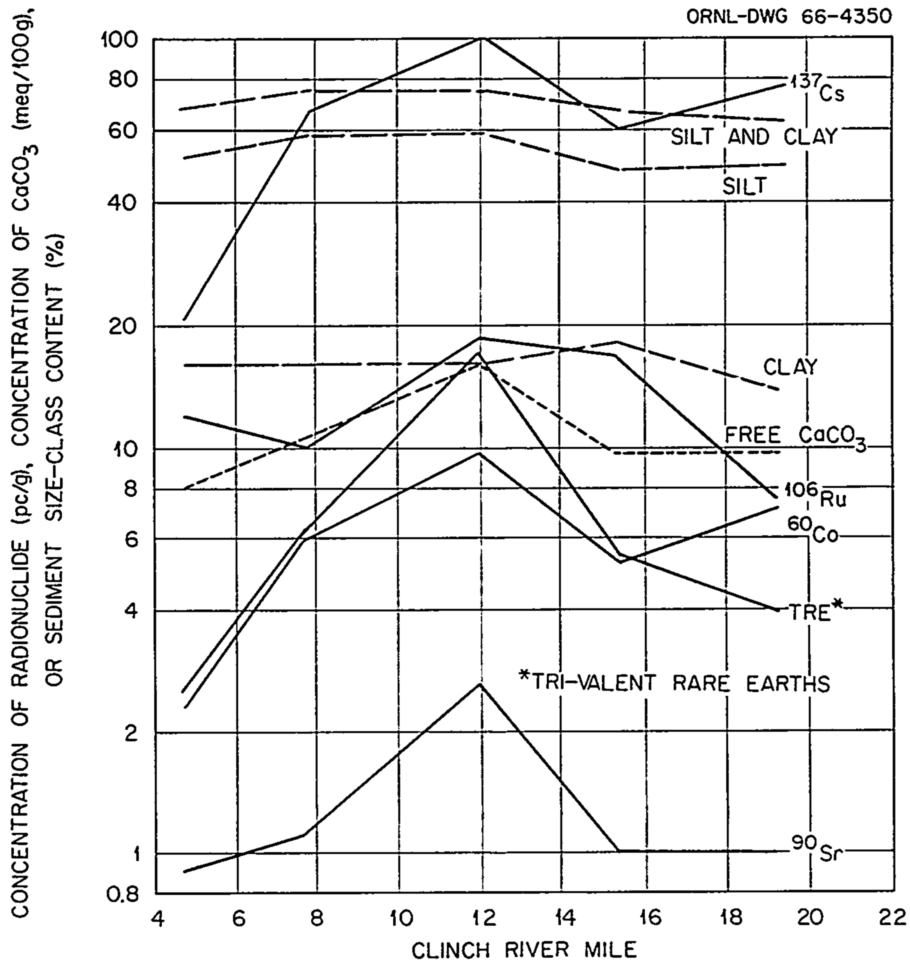

Fig. 20. Longitudinal Variation in Radionuclide Content, in Free Calcium Carbonate Content, and in Particle-Size Distribution of Clinch River Bottom Sediment

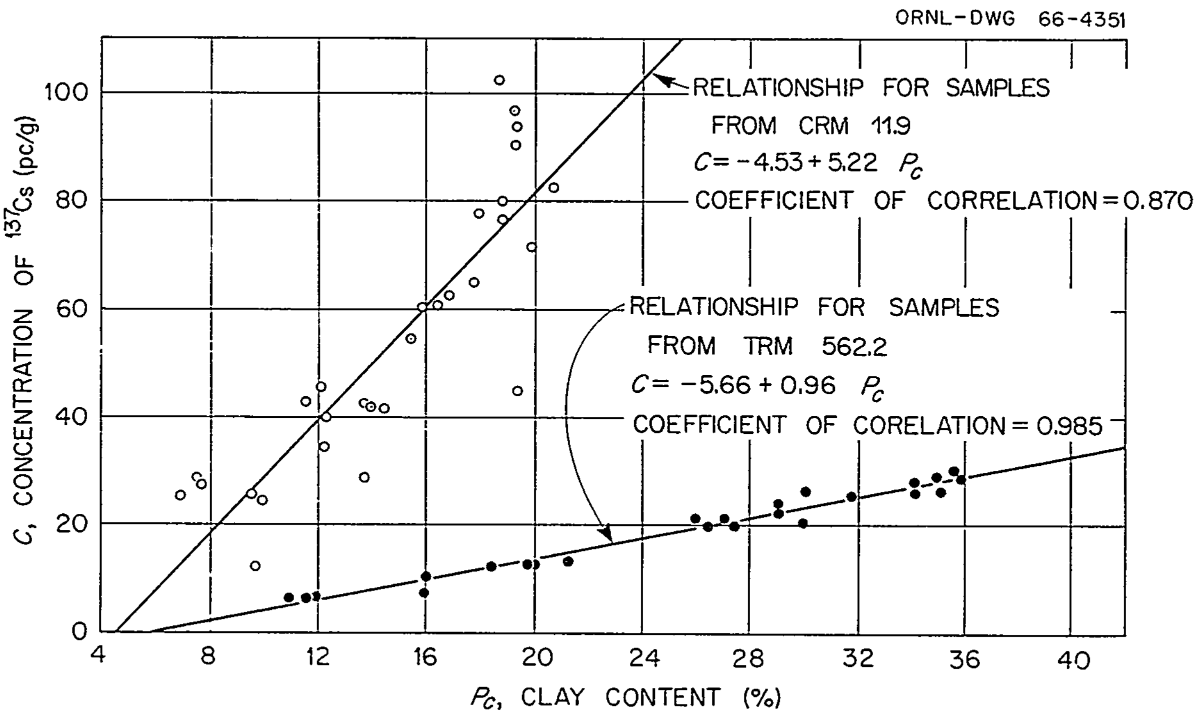

Fig. 21. Relation of Content of Clay-Size Fraction (<2μ diameter) in Bottom Sediment to Radionuclide Concentration in Sample.

Sorption and Cation Exchange Properties of Bottom Sediments

The principal means of uptake of radionuclides by Clinch River bottom sediments has been postulated to be cation exchange, a type of sorption. For this reason, the total cation exchange capacity and the exchangeable cation content of bottom sediment cores collected at the five sections sampled during 1961 were determined by the U. S. Geological Survey.

The total cation exchange capacity, determined by an ammonium chloride leaching method, was much the same for all samples (11.3 ± 0.2 meq/100 grams). The total "exchangeable" cation contents varied, however, and in all samples exceeded the determined total cation exchange capacity. Because exchangeable sodium and potassium were not present, and the content of exchangeable magnesium was almost the same in all samples (6.0 ± 0.2 meq/100 grams), the variation in total "exchangeable" cation content is obviously the result of variation in the "exchangeable" calcium content of the five samples. The anomalously high "exchangeable" calcium content, higher in all samples than the determined total cation exchange capacity, together with the moderately high pH's of the samples, indicates that calcium carbonate in the sediment was dissolved when the sediment was leached with ammonium chloride. This implication concerning the mineralogy of the bottom sediments will be discussed in a later paragraph.

In order to further define the cation exchange properties of Clinch River bottom sediments, eleven sediment samples of the composited cores collected from the Clinch River in 1960 were evaluated for their ability to sorb strontium. In demineralized water containing ^{85}Sr as a radioactive tracer (5.0×10^{-5} meq of strontium in 200 ml), a mean value of 49.3 ± 1.7 percent of the strontium was removed by 0.1 gram of the sediments. The data on exchangeable cation content of the composite samples indicate that the saturating cations on the clays are calcium and magnesium. If in a system of this sort approximately 50% of the strontium can be sorbed, leaving 50% of the strontium still in solution, then one would suspect that the calcium plus magnesium on the exchange sites equals that in solution, i.e., it equals 50% of the total calcium plus magnesium (ion exchange plus dissolved) in the system. This suspicion is based on the assumption that the ratio for Sr/Mg selectivity by clays is the same as the observed ratio for Sr/Ca selectivity by clays, i.e., 1.0.

The source of the dissolved calcium in the sediment-distilled water mixtures used in the sorption studies can be assumed to have been calcium carbonate contained in the sediment. The solubility of calcium carbonate has been reported to be 0.014 grams/liter[23]. This value would correspond to 0.056 meq Ca in 200 ml of saturated solution, which is greater than the 0.022 meq Ca that would be 50% of the total calcium plus magnesium in the system (0.022 meq Ca + Mg on exchange sites per 0.1 gram sediment and 0.022 meq Ca + Mg in solution based on total exchangeable cation content of sediment). Hence, it can be assumed that when strontium sorption was measured using demineralized water, dissolution of calcium carbonate in the sediment had not yet reached equilibrium.

When Oak Ridge tap water was substituted for demineralized water, the calcium concentration in the solution was increased to 26 ppm, and the magnesium concentration to 8 ppm, which resulted in a solution containing 0.39 meq of Ca and Mg in 200 ml. The sorption of strontium in this system was reduced to 5.07 ± 0.27 percent of that in the demineralized water system. This effect can be explained if it is assumed that the sorption of strontium by the sediment in the tap water system should be reduced in proportion to the reduction in the Sr/(Ca + Mg) ratio in the tap water system as compared to the same ratio in the distilled water system. Thus,

$$\frac{\text{Sr in distilled water system}}{\text{Ca + Mg in distilled water system}} = \frac{5 \times 10^{-5} \text{ meq}}{0.022 \text{ meq}} = 0.00114,$$

and

$$\frac{\text{Sr in tap water system}}{\text{Ca + Mg in tap water system}} = \frac{5 \times 10^{-5} \text{ meq}}{0.39 \text{ meq}} = 0.00013,$$

so reduced sorption should be $\frac{(0.00013)}{(0.00114)}$ $(49.3\%) = 5.6\%$.

Thus, the high concentration of dissolved calcium in the river water, the apparently calcium-saturated status of the sediment, and the low exchange capacity of the sediment all contribute to the very low strontium content of Clinch River bottom sediment.

Sorption of cesium by sediment samples collected during 1960 has been studied by the same methods used in the strontium sorption studies described above[24]. Sorption capacities were high in both tap and demineralized water (93.1 $\pm$ 0.25 and 97.6 $\pm$ 0.25 percent respectively), and thus provide an explanation for the high ^{137}Cs content of Clinch River bottom sediment.

Sediment samples taken at CRM 22.6, above the mouth of White Oak Creek, were composited to provide material for sorption studies made by Sorathesn, et al[25]. The mineralogical composition of the clay fraction of the sediment, determined by X-ray diffraction methods, is 60% illite, 15% kaolinite, 10-15% vermiculite, and 10-15 % quartz. The 140 mesh size fraction of the crushed sediment was used in determining its sorption capacities for the radionuclides ^{137}Cs, ^{60}Co, ^{85}Sr, and ^{95}Zr-Nb. The results of these determinations are shown in Table 7.

The high uptake of ^{137}Cs by the sediment has been ascribed

Table 7. Sorption of Radionuclides by Bottom Sediment from CRM 22.6[25,a]

Radionuclide	Contact Time	Activity Sorbed (%)		K_d[b]	
		pH 6	pH 9	pH 6	pH 9
^{137}Cs	1 hour	53.75 ± 0.42	61.31 ± 0.37	2,326	3,169
	3 days	96.17 ± 0.25	96.16 ± 0.25	50,152	50,152
	7 days	97.78 ± 0.24	97.64 ± 0.24	88,048	82,769
^{60}Co	1 hour	46.44 ± 0.33	71.91 ± 0.22	1,734	5,120
	3 days	93.34 ± 0.17	82.38 ± 0.19	28,017	9,354
	7 days	97.28 ± 0.17	85.12 ± 0.18	71,567	11,445
^{85}Sr	1 hour	21.42 ± 0.78	24.79 ± 0.67	545	659
	3 days	45.79 ± 0.36	63.87 ± 0.26	1,690	3,537
	7 days	41.83 ± 0.39	66.80 ± 0.25	1,438	4,024
^{95}Zr-^{95}Nb	1 hour	62.83 ± 0.32	54.37 ± 0.37	3,380	2,383
	3 days	82.69 ± 0.25	75.55 ± 0.27	9,554	6,181
	7 days	86.56 ± 0.24	79.94 ± 0.25	12,886	7,970

[a]0.1 gram of sediment contacted with 200 ml of master solution.

[b]K_d, the distribution coefficient, is the ratio of the fraction of ions sorbed per unit weight of clay to the fraction of ions remaining in solution per unit volume of solution.

to its high illite content. Sorption of ^{85}Sr and ^{60}Co by sediment was higher than had been expected on the basis of the sediment's mineralogy, and may have been due in part to the presence of organic material with ion exchange properties in the sediment. More recent work has shown that peat moss, plant remains extracted from river sediment, and raw river sediment all possess a strong affinity for ^{60}Co.

The effect of pH on sorption of the various radionuclides by the sediment (Table 7) is in part a reflection of the ability of the radionuclides to displace sorbed hydrogen ions. Cesium exchanges with hydrogen ions to a much greater extent than strontium, so the sorption of cesium is less adversely affected by an increase in abundance of hydrogen ions competing for ion exchange positions than is the sorption of strontium. The nuclides ^{60}Co and ^{95}Zr-Nb were present as colloidal particles in the solution, and thus were flocculated and could be centrifuged out more effectively at pH 6 than at pH 9 where the hydroxyl ions apparently helped stabilize the negatively charged colloidal particles.

Mineralogy of Bottom Sediments

A knowledge of bottom-sediment mineralogy is requisite to an understanding of radionuclide distribution in the sediment because of differences between minerals in both total sorption capacities and capacities for sorbing specific nuclides. Most clay minerals and certain oxides are known to have high total cation adsorption capacities; vermiculite with a particular lattice spacing perferentially sorbs cesium[27].

Cross-sectional composites of bottom sediment cores collected in 1961 (CRM 4.7, 7.6, 11.9, 15.3, and 19.2) were examined by the U. S. Geological Survey for variations in mineralogy of the sand-, silt-, and clay-size fractions. The sand-size fraction of the sediment is composed almost entirely of quartz grains with traces of feldspar and dolomite. The silt-size fraction also consists almost entirely of quartz grains, with traces of feldspar, dolomite, and possibly diaspore. Quartz is the predominant mineral in the clay-size fraction, but appreciable quantities of clay minerals are also present; proportions of minerals in clay-size fraction are: 30-40% quartz, 20% vermiculite, 20% mica, 10-20% kaolinite, and 10% randomly interstratified vermiculite-mica.

Any comparison of radionuclide content in the 1961 core samples with their mineralogy must be very general because of the semiquantitative nature of the mineral determinations. Of the three relative mineral content variations mentioned, the variation in mica content of the samples might be expected to have the most pronounced effect on radionuclide sorption. Without more precise mineralogic data, however, little more can be said than that differences in the mica contents of the composites might be in part responsible for differences in their radionuclide contents. In general, the composite sediment samples for all five cross sections sampled have much the same mineralogy. This fact may explain the lack of variation in the total cation exchange capacities of the five samples.

The calcium carbonate content of each composite sample was estimated from the exchangeable cation content and total cation exchange capacity of the sediment as follows:

$$(CaCO_3) = Ca_X^{++} - R_T^{++} + Mg_X^{++}$$

in which

$(CaCO_3)$ = calcium carbonate in sediment in meq/100g,

Ca_X^{++} = exchangeable calcium in sediment in meq/100g,

R_T^{++} = total cation exchange capacity of sediment in meq/100g, and

Mg_X^{++} = exchangeable magnesium in sediments in meq/100g; or

$CaCO_3$ = exchangeable Ca minus total cation exchange capacity plus exchangeable Mg.

This calculation was made on the assumption that the amount of calcium held on mineral exchange sites is equal to the difference between the total cation exchange capacity and exchangeable magnesium.

Because exchangeable sodium and potassium were shown to be absent, it was assumed that no other exchangeable cations were present. The amount of "exchanged" calcium (from analysis) in excess of the estimated calcium on exchange sites is thus assumed to have been contributed by free calcium carbonate in the sample. Variations in the calcium carbonate content with distance are very similar to variations in ^{90}Sr concentrations, as is demonstrated in Fig. 20, and may indicate a direct association of ^{90}Sr in bottom sediments with free calcium carbonate.

The nearly identical variations with distance of cesium and cobalt concentrations in the composite samples suggest that those two nuclides are incorporated into the bottom sediment by the same, or similar, processes.

Discussion.-- Several shortcomings must be recognized in connection with the determinations to date of the physical properties and mineralogy of Clinch River bottom sediments. Mineral determinations were made by X-ray diffraction only, a method which cannot be expected to detect amorphous substances such as organic fragments. Coal has been observed to be fairly abundant in the sand-size fraction of sediments collected near the mouth of Poplar Creek and in certain downstream locations, but its relative abundance is not known because petrographic examinations of the bottom sediment samples were not made. An analysis by the USPHS of a leaf fragment collected at the mouth of the Emory River showed that the organic leaf material contained forty times as much radioactivity per gram as did clayey silt from the same location[28], yet little data are available on plant and animal detritus in the sediment. Also, care must be taken to avoid dissolution of highly soluble chemical constituents during the processing of sediment samples.

RADIATION DOSAGE FROM BOTTOM SEDIMENT

Main Channels of Clinch and Tennessee Rivers

Re-examination of Figs. 9 and 10 is useful in gaining an impression of reaches in which high gamma radiation dosage may occur. As mentioned in the section on annual monitoring surveys the highest radiation levels occur in the Clinch River; and the highest levels in the Clinch River occur between CRM 5 and 15. Data for curves in these two figures are average count rates in the sections. As pointed out in the section "Influence of Dispersion and Flow Pattern" count rates vary across the sections, with a likelihood of the maximum count rate being considerably different from the average in the Clinch River. The pattern of longitudinal distribution of maximum count rate is generally similar to the pattern for average count rate. Hence, the aforementioned curves for average count rates delineate reaches that would be of major interest in a safety analysis.

The maximum count rate for the period of record (1954-61) was observed in 1959 at CRM 15.2 on the Clinch River: 394 cps (average, 176 cps); maximum count rate observed in 1961 was at CRM 5.8: 219 cps (average, 137 cps). As pointed out by Cottrell[6] and by Cowser and Snyder[16] these maxima were well below maximum permissible dose rates even if the exposure to radiation from the sediments were continuous.

Estimates of gamma radiation dosages, using flounder count rates, are meaningful only upstream from Chattanooga, Tenn. Garner and Kocktitzky[5] have shown that the radiation due to fission products in bottom sediments cannot be distinguished from radiation due to naturally occurring radionuclides downstream from this city. Cowser and Snyder[16] have investigated the relation between gamma radiation dose and flounder count rate and have concluded that such relationships are particularly meaningful only in the Clinch River.

Water shields radiation emitted from the sediments; 0.7 foot of water attenuates the dose by a factor of about two. Hence, the most critical situation, in general, for external exposure to this radiation will occur if the person is in direct contact with the contaminated sediment, sitting or lying on the sediment. This type of exposure can occur under at least two conditions: (1) the unusual circumstance of divers working on the riverbed, and (2) people sitting or lying on the river banks during 'winter-flow' conditions (fishermen). During the winter, water levels in Watts Bar Lake are about six feet lower than during the summer. As a result sediments in contact with radioactive waters during the summer are exposed. The enveloping curve in Fig. 22 gives evidence that the maximum flounder count rate on the exposed riverbank in the winter of 1961 would have been 105 cps, about one-half of the maximum count rate observed in the 1961 annual monitoring survey.

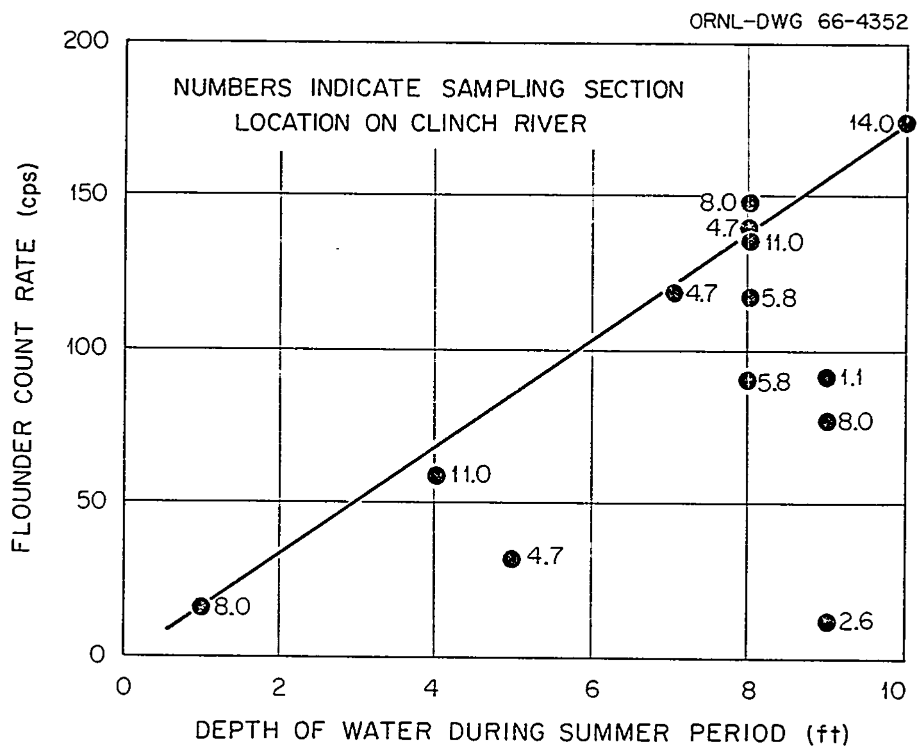

Fig. 22. Estimate of Maximum Flounder Count Rate on Exposed Banks of Clinch River when Lake is Drawn Down

Clinch River Sloughs

In 1963, a survey of radioactivity in bottom sediment of six large sloughs of the Clinch River and one slough of the Emory River was made. The purpose of the survey was to determine the level of surface radioactivity in fine sediment of the sloughs. Radioactivity readings were made with the Flounder instrument.

The results of the survey indicate that, though the radioactivity of sediment in the sloughs is within the range of radioactivity in the main river channel, the maximum radioactivity value was not as high as the maximum radioactivity in adjacent portions of the main channel[4].

In several of the deeper sloughs, a direct relationship between bottom sediment radioactivity and depth of water was found. Causes of this relationship are not definitely known. Two erosional processes may be largely responsible: (1) subaerial erosion of exposed sediments during winter low water in Watts Bar Lake, and (2) movement of sediment from shallower to deeper portions of the sloughs by sheet erosion[4].

CONCLUSIONS

Contents of this report illustrate the wealth of information available which is related to association of radioactivity with bottom sediments of the Clinch and Tennessee Rivers. Action by the Clinch River Study Steering Committee which catalyzed study of this information was well taken. Through compilation and interpretation of pertinent data, perspective has been gained as to how well investigations have met objectives of the Study (p. 4) and to suggest direction of other investigations, such as those reported in Part B of this report.

The first objective of the Clinch River Study is "to determine the fate of radioactive materials currently being discharged to the Clinch River".

The predominant radionuclide retained in bottom sediments, determined in the inventory of radioactivity of the uppermost strata of bottom sediments of the Clinch River, is ^{137}Cs. By application of an appropriate scaling factor to this inventory of the ^{137}Cs content in the sediments, probable retention of releases of this radionuclide to the study reach of the Clinch River is estimated to be 20 percent. Continued application of the same scaling factor to this incomplete inventory suggests that the fraction of retention for other radionuclides released to the Clinch River is much less than for ^{137}Cs, especially for ^{90}Sr (almost negligible).

Results of this partial inventory suggest only a small fraction of the fission-product load released from White Oak Lake has been retained in bottom sediments. Work was specifically undertaken to accurately determine fission-product activity retained in the bottom sediments, and results of this work are described in Part B of this report.

Several factors may influence deposition and retention of radioactive materials in bottom sediments of the Clinch and Tennessee Rivers. Among these factors are variations in radionuclide loads released from White Oak Lake, diffusion, sedimentation, some aspects of flow pattern, particle size, sorption, mineralogy, and states of calcium equilibrium in water and sediment. Other factors that appear to be important, but which are not reported in this paper, are plant and animal detritus contents of sediment, coal content, potential for desorption of radionuclides (see Morton[4]), and the physico-chemical state of radionuclides in water and in suspended and deposited sediments (see Part B, this report).

Study of these factors has a direct bearing on satisfying the aims of the second objective of the study: "to determine and understand mechanisms of dispersion...".

Evidence has been obtained that changes in the concentration of radionuclides at the surface of bottom sediments are directly related to variations in radionuclide loads released through White Oak Dam. Evidence has also been obtained that suggests that most radionuclides that are associated with bottom sediment are sorbed on the sediments, or are in the form of particulate matter, prior to their release to the river (as suspended sediment) and that sedimentation is the primary mechanism for incorporation of radionuclides in bed deposits.

Effects of thermal stratification on depositional patterns of sediments in the Clinch River do not seem to be of primary importance. Other hydraulic characteristics, however, do affect depositional patterns: diffusion of sediment-bearing water from White Oak Creek into the Clinch River, surface area of the channel bed, and flow pattern.

In at least the first three miles downstream from the mouth of White Oak Creek lateral distribution of contaminated sediments is constrained, being related to the diffusion of White Oak Creek waters in the river. Between 4 and 6 miles downstream from the creek mouth these waters become fully mixed in the river[1, 3, 18] and lateral distribution of contaminated sediments should be independent of this particular aspect of diffusion.

As radioactive material is dispersed over a larger and larger area, concentration of radionuclides in bottom sediments is decreased.

Deposition of sediments and lateral distribution of radioactivity in these sediments are influenced by flow patterns around bends and about islands. Greater deposition and higher concentrations occur in eddy zones. Attempts to quantitatively relate concentration to curvature of bends, although not brought to complete fruition, are encouraging.

Sorption of radionuclides in river sediments may be expected to vary with changes in particle size, because the relative mineral composition usually varies with particle size and the chemically reactive surface of the particle increases, per unit weight, as its size decreases. Such concepts have been tentatively confirmed for sediments in the Tennessee River System by directly relating concentration of radionuclides to clay content of the sample. In the Clinch River the variation in radionuclide contents of five samples, collected at widely spaced (longitudinally) sections, was found to be greater than variations in particle size; essentially this contradicts findings for the samples from the Tennessee River and must for the moment remain an unexplained anomaly.

Only sorption of strontium and cesium by bottom sediments of the Clinch River has been measured extensively. Sorption of both radionuclides was found to be practically constant with longitudinal location. In tracer studies, using bottom sediments, sorption of strontium was found to be 49.3 ± 1.7 percent and for sorption cesium it was 97.6 ± 0.25 percent in demineralized-water solutions.

High sorption of ^{137}Cs has been ascribed to high illitic-type mineral content of the sediments. An appreciable content of illitic-type minerals, particularly in the clay-size fraction, is in the riverbed. Mineralogic composition of these samples is fairly constant. Sand and silt sizes have consisted mostly of quartz. Mineral composition of clay-size particles is more varied than for large sizes; variations in content of one mineralogic group in this size fraction, the mica-related minerals, may be the most important control on cesium sorption in the sediments.

Similar variations in ^{90}Sr content of the riverbed sediments and in free calcium content have been noted in the Clinch River. This relationship may indicate that calcium in the water and sediment has a considerable influence on association of ^{90}Sr with bottom sediment.

Studies of physico-chemical properties of bottom sediments in the Clinch River have not shown any property which may singularly affect variations in radionuclide content of these sediments. Incorporation of radionuclides in sediments is due to complex interaction of a number of properties: size distribution, mineralogy, sorption capacities, and cation exchange capacities.

Desorption of radionuclides from riverbed sediments is being studied. Progress on investigations by Bonner and Tamura has been reported by Morton[4].

Geochemical studies, needed to delineate the processes which lead to incorporation of radionuclides in bottom sediment, strongly suggest that radioactivity has become associated with suspended sediments in White Oak Lake and that incorporation of some radioactivity in sediments on beds of the Clinch and Tennessee Rivers is the result of a hydraulic process, sedimentation. The possibility exists that some of the radioactivity is not associated with lithologic or biotic sediments; this radioactivity may have become associated with bed deposits through formation of chemical precipitates and subsequent agglomeration of these precipitates into particles of settleable size. Another possibility exists that these precipitates are occluded with other settleable-sized sediments and are thereby included into bed material.

Parts of material presented in this report are germane to safety analysis and will serve as an aid to achieving the third objective of the Study: "to evaluate direct and indirect hazards of current disposal practices...".

Highest levels of radiation and highest concentration of radionuclides are found to occur in bottom sediments of the Clinch River rather in reaches of the Tennessee River. Higher concentrations of radionuclides are found to occur in the reach between CRM 5 and 15 than elsewhere in the Clinch River.

The dominant radionuclide in these sediments is ^{137}Cs; very little ^{90}Sr is present.

Although there have been exceptions, contamination of the bottom sediments, due to release of radioactivity from White Oak Lake, is negligible upstream from the mouth of White Oak Creek in the Clinch River and upstream from the mouth of the Clinch River in the Tennessee River. Contamination of bed deposits in the Tennessee River has been found as far downstream as the Ohio River (Kentucky Dam). From information obtained in 1961, contamination to the mouth of the Tennessee River was limited to ^{106}Ru. However, releases of ^{137}Cs, ^{90}Sr, ^{60}Co, and ^{144}Ce were considerably less in 1961 than in previous years. Considering that concentration of radionuclides in bottom sediment is related to load of radionuclides released through White Oak Da, it appears possible that higher releases in years prior to 1961 could have produced contamination of sediments in reaches much farther downstream than those observed in 1961, possibly to the mouth of the Tennessee River for radionuclides other than ^{106}Ru.

Radiation levels of bottom sediments in sloughs of the Clinch River are not as high as those for sediments on the bed of the main stream. In addition, radiation levels of sediments deposited on parts of the slough or riverbed that are above 'winter' water levels are considerably less than levels measured at other locations which are continuously inundated.

Most radiation detecting devices used to measure distribution of radioactivity in bottom sediments in situ have performed well in the Clinch River. Farther downstream in the Tennessee River the higher levels of natural radiation and radiation from fallout relative to radiation from fission-product contamination originating from ORNL tend to limit the usefulness of these detectors. One limitation of detectors used is that the instruments do not have the capability to discriminate radiation emitted from one radionuclide from that emitted by another.

An attempt at this time to evaluate the usefulness of this river system for the disposal of radioactive materials, the fourth objective of the Study, would seem to be premature insofar as bottom sediments are concerned. Lack of an inventory, an incomplete picture of the relative importance of various physical and chemical factors which lead to the association of fission products with sediments in channel beds, and only the beginnings of work on desorption mechanisms have led to this conclusion of prematurity.

Technologically, results of investigations reported in this paper may be helpful in making suggestions towards long-term monitoring procedures, the fifth and final objective of the Study. The Subcommittee on Bottom Sediment Sampling and Analyses made recommendations as outlined in the following section of this report to the Clinch River Study Steering Committee on December 11, 1964. Many of the suggestions stem from interpretation in this report of work by personnel of the Applied Health Physics Section, ORNL.

Early recognition of problems involved in monitoring radioactivity and continued responsible action by personnel of the Applied Health Physics Section provided abundant data with which to make assessments of fission-product content of bottom sediments in Tennessee River reservoirs.

1.: Monitoring of the radioactivity in the bottom sediments in the Clinch and Tennessee Rivers should be continued as long as radioactive materials are released to the Clinch River, and for a limited time thereafter. The present bottom sediment monitoring program extending from the mouth of White Oak Creek on the Clinch River to the mouth of the Tennessee River should be maintained.

Studies by the Applied Health Physics Section, ORNL indicate that radioactivity in bottom sediments above background levels has moved to sections in the Tennessee River progressively farther downstream from the mouth of White Oak Creek. Some information on radioactivity in bottom sediments should be collected as long as releases of radioactive materials to the Clinch River from White Oak Lake, or other sources at ORNL, are continued. Radioactivity stored in the seepage pits and trenches, in the burial grounds, and in the sediments and soils of White Oak Creek Basin may continue to be released from White Oak Lake for a period of time after cessation of all controlled releases to the basin. It is suggested that the monitoring continue after such cessation until such time as the level of radioactivity in releases from White Oak Creek has dropped below a prescribed limit of concentration.

2.: The primary stations in a monitoring network should be the water sampling stations at White Oak Dam (or other control works near the mouth of White Oak Creek) and at the tailrace of Melton Hill Dam (or other upstream stations below Melton Hill Dam).

Studies by personnel of this subcommittee indicate that changes in the gamma radioactivity of the bottom sediments are directly releated to changes in the load of radionuclides released from White Oak Lake. The contribution to the total load of some radionuclides in the study reaches, especially strontium-90, upstream of the mouth of White Oak Creek is appreciable. If the radionuclide releases from White Oak Lake continue to decrease, the proportion of this upstream contribution to the total load will become more significant.

3.: Present procedures for the monitoring of radioactivity in the bottom sediments used by the Applied Health Physics Section, ORNL, are satisfactory. A general pattern of longitudinal distribution of radioactivity has been found in the past several years. After operations at Melton Hill Dam are begun, a new pattern may develop. Surveys should be made to determine when this new pattern of longitudinal distribution has been established. If power releases from Melton Hill Reservoir are affecting the distribution of radioactivity, it should be the responsibility of the Applied Health Physics Section to make appropriate modifications in the procedures to document the effects of these power releases.

At present the procedures include the determination of the level of radiation at the surface of the sediment and the determination of the concentration of radionuclides in composite samples of sediments at the surface of the deposits. These methods should not be considered standard because newer techniques in determination of radiation levels and of radionuclide concentration may indicate that the procedures should be up-dated.

Modifications in the frequency of surveying radioactivity in bottom sediments and the selection of sections for such surveys may be suggested by information presented in this report.

The determination of bed profiles at sections which are monitored is suggested so as to provide additional information for interpretation of radiation and radionuclide concentration data.

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APPENDIX

Tables 8 through 20

Table 8. Concentration of Cesium-137 in Upper Portion of Bottom Sediments of Clinch River, 1954–61[a]

Location (CRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
21.5	3	5		5	4	4.5	1.5	1.3
19.1	12	7	116	528	44	527	64	41
16.3	27	22	208	177	223	464	175	71
15.2	22	34	268	119	146	391	374	64
14.0	24	29	115	184	298	464	257	127
11.0	22	34	144	251	236	229	240	98
8.3[b]	22	38	244	178	170	236	194	81
5.7[c]	24	29	266	299	223	207	177	115
4.7	22			236	151	169	176	112
2.6	15			173	92	171	93	82
1.1	24	25	257	192	67	259	141	100

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]CRM 8.0 beginning in 1959.
[c]CRM 5.8 beginning in 1959.

Table 9. Concentration of Cesium-137 in Upper Portion of Bottom Sediments of Tennessee River, 1954–61[a]

Location (TRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
				Fort Loudoun Reservoir				
604.1	2	2	5		2			1.2
								0.99
				Watts Bar Reservoir				
570.8	3			5	2	1.8	1.5	1.1
562.7	10	7	73	55	51	41	46	34
552.7	12			57	36	41	32	42
534.8	5			47	22	23	28	15
532.0	10	11	32	39	21	26	24	25
				Chickamauga Reservoir				
509.5	3			20	10	6.8		1.1
491.9	5		20	20	16	14	18	14
475.1[b]	5	2	14	16	13	11	11	11
				Hales Bar Reservoir				
434.1				13	9	13	10	4.5
				Guntersville Reservoir				
381.2				7	7	3.2	6.3	3.5
354.5[c]				7	4	3.6	4.0	4.6
				Wheeler Reservoir				
337.6								0.45
320.9								2.5
280.0								4.6
				Wilson Reservoir				
267.4								4.8
261.3								6.9
				Pickwick Landing Reservoir				
227.4								2.0
207.3								2.6
				Kentucky Reservoir				
97.2								1.1
75.7								0.99
67.0								0.90
45.8								0.99
24.6								1.2

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]TRM 471.7 beginning in 1960.
[c]TRM 354.4 beginning in 1959.

Table 10. Concentration of Strontium-90 in Upper Portion of Bottom Sediments of Clinch River, 1954–61[a]

Location (CRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
21.5	1			1	1	0.5		0.26
19.1	5		4	3	2	9.5	0.7	1.0
16.3	5	4	7	5	6	9.0	2.3	2.0
15.2	5		9	5	6	7.2	4.4	0.77
14.0	5	4	4	3	11	8.6	2.7	1.1
11.0	5	4	6	5	13	5.4	3.7	1.0
8.3[b]	4	4	6	5	6	5.9	3.5	1.4
5.7[c]	4	4	6	7	1	4.5	2.6	1.0
4.7	4			5	8	4.5	1.4	1.3
2.6	3			3	5	1.4	1.0	0.90
1.1	4	3	6	3	5	2.7	1.4	0.41

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]CRM 8.0 beginning in 1959.
[c]CRM 5.8 beginning in 1959.

Table 11. Concentration of Strontium-90 in Upper Portion of Bottom Sediments of Tennessee River, 1954–61[a]

Location (TRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
				Fort Loudoun Reservoir				
604.1	2	1.4	1.3		1.1			0.14
								0.18
				Watts Bar Reservoir				
570.8	2			0.9	1.0	0.5	0.2	0.14
562.7	2	0.3	3	0.8	2.0	0.9	0.5	0.49
552.7	2			0.5	1.5	1.4	0.5	0.59
534.8	2			0.9	1.7	0.9	0.5	0.27
532.0	4	0.4	3	0.6	1.7	1.4	0.7	0.63
				Chickamauga Reservoir				
509.5	3			1	1.6	0.5		0.27
491.9	2		2	0.6	0.1	0.9	0.9	0.36
475.1[b]	2	0.3	2	1.3	1.3	0.9	0.4	0.36
				Hales Bar Reservoir				
434.1				1.4	1.2	0.9	0.5	0.36
				Guntersville Reservoir				
381.2				0.8	1.9	0.5	0.5	0.72
354.5[c]				0.7	1.5	0.5	0.5	0.36
				Wheeler Reservoir				
337.6								0.32
320.9								0.09
280.0								0.36
				Wilson Reservoir				
267.4								0.54
261.3								0.45
				Pickwick Landing Reservoir				
227.4								0.31
207.3								0.23
				Kentucky Reservoir				
97.2								0.45
75.7								0.18
67.0								0.23
45.8								0.18
24.6								0.41

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]TRM 471.7 beginning in 1960.
[c]TRM 354.4 beginning in 1959.

Table 12. Concentration of Cerium-144 in Upper Portion of Bottom Sediments of Clinch River, 1954–61[a]

Location (CRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
21.5	2	4		5	12	6.8	0.8	0.44
19.1	5	6	24	33	7	92	9.0	2.7
16.3	8	21	37	12	20	71	23	5.6
15.2	7	32	56	9	22	54	39	4.4
14.0	8	22	20	7	43	65	25	8.2
11.0	8	31	41	10	40	27	34	9.4
8.3[b]	5	32	48	10	16	30	25	9.4
5.7[c]	8	40	56	12	24	20	22	9.9
4.7	7			13	21	18	18	9.9
2.6	4			9	17	9	11	7.0
1.1	5	30	44	13	22	20	11	8.6

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]CRM 8.0 beginning in 1959.
[c]CRM 5.8 beginning in 1959.

Table 13. Concentration of Cerium-144 in Upper Portion of Bottom Sediments of Tennessee River, 1954–61[a]

Location (TRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
				Fort Loudoun Reservoir				
604.1	1	1.7	3		4.7			0.77
								0.99
				Watts Bar Reservoir				
570.8	1			1.3	5.7	6.8	0.9	0.72
562.7	2	13	15	5.5	8.0	92	3.1	2.9
552.7	2			4.3	9.6	71	1.8	2.9
534.8	1			3.0	7.2	54	2.3	1.1
532.0	2	15	8	2.6	4.9	65	1.8	2.3
				Chickamauga Reservoir				
509.5	1			1.9	6.2	27		0.23
491.9	2		6	1.8	4.6	30	1.8	1.8
475.1[b]	2	4	4	1.6	6.2	20	1.0	1.5
				Hales Bar Reservoir				
434.1				3.4	7.2	18	1.6	1.1
				Guntersville Reservoir				
381.2				3.4	5.4	9.0	1.3	0.76
354.5[c]				1.6	4.7	20	0.6	0.95
				Wheeler Reservoir				
337.6								0.27
320.9								0.77
280.0								1.5
				Wilson Reservoir				
267.4								1.8
261.3								2.4
				Pickwick Landing Reservoir				
227.4								0.90
207.3								0.99
				Kentucky Reservoir				
97.2								0.54
75.7								0.68
67.0								0.50
45.8								0.86
24.6								0.86

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]TRM 471.7 beginning in 1960.
[c]TRM 354.4 beginning in 1959.

Table 14. Concentration of Trivalent Rare Earths[a] in Upper Portion of Bottom Sediments of Clinch River, 1954–61[b]

Location (CRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
21.5	1	3		2	3	5	1.4	0.7
19.1	2	3	7	10	6	251	23	7.8
16.3	4	5	11	5	13	151	74	16
15.2	4	7	15	4	17	142	144	16
14.0	4	8	7	4	21	149	132	31
11.0	6	16	19	8	18	58	177	29
8.3[c]	4	24	19	6	14	71	144	26
5.7[d]	8	12	18	7	15	57	69	28
4.7	5			6	13	33	38	20
2.6	5			5	10	19	72	23
1.1	4	9	15	5	12	41	48	35

[a]Analysis includes ^{90}Y as a constituent but excludes ^{144}Ce.
[b]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[c]CRM 8.0 beginning in 1959.
[d]CRM 5.8 beginning in 1959.

Table 15. Concentration of Trivalent Rare Earths[a] in Upper Portion of Bottom Sediments of Tennessee River, 1954–61[b]

Location (TRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
				Fort Loudoun Reservoir				
604.1	2	1.7	3		4.8			0.86
								1.4
				Watts Bar Reservoir				
570.8	1			1.1	5.1	1.7	0.5	1.1
562.7	3	6	6	1.9	5.5	8.3	21	8.2
552.7	1			2.7	6.1	7.0	12	5.7
534.8	2			1.3	5.5	5.4	8.6	3.1
532.0	4	7	4	1.5	5.5	6.6	5.1	4.0
				Chickamauga Reservoir				
509.5	3			1.7	6.1			1.2
491.9	2		3	1.3	5.3	2.9	5.0	3.2
475.1[c]	2	6	1.8	1.0	6.4	5.5	3.1	3.4
				Hales Bar Reservoir				
434.1				1.8	8.1	7.0	3.0	1.5
				Guntersville Reservoir				
382.1				1.3	2.6	0.2	0.9	1.4
354.5[d]				1.4	4.7	2.5	2.0	1.9
				Wheeler Reservoir				
337.6								0.54
320.9								0.86
280.0								0.68
				Wilson Reservoir				
267.4								1.6
261.3								1.8
				Pickwick Landing Reservoir				
227.4								0.72
207.3								1.0
				Kentucky Reservoir				
97.2								0.86
75.7								0.32
67.0								1.1
45.8								0.77
24.6								0.54

[a]Analysis includes ^{90}Y as a constituent but excludes ^{144}Ce.
[b]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[c]TRM 471.7 beginning in 1960.
[d]TRM 354.4 beginning in 1959.

Table 16. Concentration of Ruthenium-106 in Upper Portion of Bottom Sediments of Clinch River, 1954–61[a]

Location (CRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1951
21.5	1			3	6	5.0	1.8	2.7
19.1	8		5	14	3	18	27	95
16.3	5	4	8	6	7	17	80	159
15.2	5		11	3	6	18	83	148
14.0	6	4	6	4	16	17	99	153
11.0	2	5	7	6	12	9.9	99	144
8.3[b]	5	4	10	5	7	11	85	152
5.7[c]	5	8	8	6	11	7.7	90	157
4.7	5			5	10	7.2	79	148
2.6	5			4	6	6.3	72	103
1.1	3	4	10	6	10	9.9	55	141

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]CRM 8.0 beginning in 1959.
[c]CRM 5.8 beginning in 1959.

Table 17. Concentration of Ruthenium-106 in Upper Portion of Bottom Sediments of Tennessee River, 1954–61[a]

Location (TRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
				Fort Loudoun Reservoir				
604.1	1	0.5	3		4.6			1.9
								2.3
				Watts Bar Reservoir				
570.8	3			1.3	2.6	3.6	2.7	2.7
562.7	2	3	4	3.1	4.1	6.3	19	34
552.7	1			3.4	5.4	5.0	17	39
534.8	2			3.1	3.1	3.6	14	22
532.0	1	4	3	2.0	2.0	3.2	13	31
				Chickamauga Reservoir				
509.5	1			2.3	3.4			8.6
491.9	1		2	1.8	3.7	3.6	27	30
475.1[b]	1	1	3	1.5	3.5	5.4	10	26
				Hales Bar Reservoir				
434.1				2.9	3.5	8.6	32	28
				Guntersville Revervoir				
381.2				0.9	2.5	2.7	12	14
354.5[c]				1.7	2.3	4.1	6.9	13
				Wheeler Reservoir				
337.6								4.1
320.9								11
280.0								16
				Wilson Reservoir				
267.4								18
261.3								25
				Pickwick Landing Reservoir				
227.4								8.7
207.3								12
				Kentucky Reservoir				
97.2								7.2
75.7								6.4
67.0								5.0
45.8								5.3
24.6								4.4

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]TRM 471.7 beginning in 1960.
[c]TRM 354.4 beginning in 1959.

Table 18. Concentration of Cobalt-60 in Upper Portion of Bottom Sediments of Clinch River, 1954–61[a]

Location (CRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
21.5	3	2			3	0.9	0.5	0.32
19.1	11		26	30	4	69	8.2	5.9
16.3	19	18	39	15	21	53	19	11
15.2	19		59	14	9	41	35	10
14.0	19	23	29	17	16	53	27	14
11.0	19	25	37	15	15	26	25	14
8.3[b]	23	29	50	15	17	28	22	11
5.7[c]	31	26	52	18	17	29	22	14
4.7	27			15	14	26	21	15
2.6	19			13	9	18	12	11
1.1	23	21	46	16	13	20	16	12

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]CRM 8.0 beginning in 1959.
[c]CRM 5.8 beginning in 1959.

Table 19. Concentration of Cobalt-60 in Upper Portion of Bottom Sediments of Tennessee River, 1954–61[a]

Location (TRM)	Concentration (pc/g)							
	1954	1955	1956	1957	1958	1959	1960	1961
				Fort Loudoun Reservoir				
604.1	4	0	1.0		0.6			0.36
								0.32
				Watts Bar Reservoir				
570.8	4			1	0.8	0.9	0.3	0.3
562.7	8	7	11	6	5.7	6.8	6.0	4.5
552.7	6			6	6.1	8.1	5.0	5.3
534.8	7			5	3.6	5.0	4.7	2.5
532.0	7	13	7	3	2.9	3.6	4.1	3.9
				Chickamauga Reservoir				
509.5	4			2	2.1			0.6
491.9	5		4	3	3.1	3.6	3.6	3.0
475.1[b]	5	4	6	3	1.7	2.7	2.3	2.4
				Hales Bar Reservoir				
434.1				2.0	1.7	2.7	2.7	1.3
				Guntersville Reservoir				
381.2				2.0	1.7	1.4	1.4	1.0
354.5[c]				0.3	2.5	1.8	1.4	1.1
				Wheeler Reservoir				
337.6								0.23
320.9								0.41
280.0								0.90
				Wilson Reservoir				
267.4								0.90
261.3								1.2
				Pickwick Landing Reservoir				
227.4								0.72
207.3								0.81
				Kentucky Reservoir				
97.2								0.45
75.7								0.41
67.0								0.32
45.8								0.41
24.6								0.41

[a]Data from annual monitoring surveys of Applied Health Physics Section, ORNL.
[b]TRM 471.7 beginning in 1960.
[c]TRM 354.4 beginning in 1959.

Table 20. Particle-Size Distribution in Bottom Sediments from Clinch River

(percent)

Location (CRM)	Clay (<2 μ)			Silt (2–62 μ)			Sand (>62 μ)		
	Lab. A	Lab. B	Average	Lab. A	Lab. B	Average	Lab. A	Lab. B	Average
4.7	13[a]	18.1	16	55[a]	49.4	52	32[a]	31.3	32
7.6	13.5[a]	17.9	16	58[a]	59.0	58	28.5[a]	21.8	25
11.9	14	18.4	16	55	62.4	59	31	19.2	25
15.3	16	20.9	18	46	50.4	48	38	28.7	33
19.2	12	16.9	14	48	49.4	49	40	33.8	37

[a]Average of duplicate samples.

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P. H. Carrigan, Jr.
R. J. Pickering

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HEALTH PHYSICS DIVISION

RADIOACTIVE MATERIALS IN BOTTOM SEDIMENT OF CLINCH RIVER: PART B, INVENTORY AND VERTICAL DISTRIBUTION OF RADIONUCLIDES IN UNDISTURBED CORES

Supplement No. 2B to Status Report No. 5 on Clinch River Study

By

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Final Progress Report, Part B, of Subcommittee on Bottom Sediment Sampling and Analysis

MARCH 1967

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

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during the coring operations and used to obtain increased recovery of cores consisting of rather fluid sediment. Contractual arrangements for drilling were under the supervision of F. L. Parker, Section Chief, Radioactive Waste Disposal Section, ORNL.

ABSTRACT

Inventory, retention, and distribution of radionuclides in bottom sediment of the Clinch River, and factors influencing association of radionuclides with this sediment have been experimentally investigated as part of the Clinch River Study. Experimental work included a field sampling program and a variety of physicochemical analyses in the laboratory.

Core samples of bottom sediment were collected by means of a Swedish Foil Sampler from a 21-mile reach of the Clinch River. The study reach extended from the mouth of the river to the mouth of White Oak Creek and included fourteen sampling sections. Four to thirteen cores were obtained from each section, the number depending on the transverse distribution of the bottom sediment.

Physicochemical analyses performed on sediment samples included gross gamma counting, gamma spectrometry, radiochemical extractions and beta counting, particle-size analyses, mineralogical analyses and determinations of cation exchange capacities and leachable cation content.

The total quantities of the principal radionuclides contained in bottom sediment in the 21-mile study reach at the time of sampling (July 1962) were: 150 curies of ^{137}Cs, 18 curies of ^{60}Co, 16 curies of ^{106}Ru, 2.9 curies of ^{90}Sr, and at least 10 curies of rare earths. Through comparison of the inventory to the total release of each radionuclide to the Clinch River for the period of record, 1943-62, (adjusting for radioactive decay) the percentage of retention is computed to be: 21 percent for ^{137}Cs, 9 percent for ^{60}Co, 0.4 percent for ^{106}Ru, and 0.2 percent for ^{90}Sr. Retention of rare earths may approach, and possibly exceeds, 25 percent.

Ninety-five percent of the total amount of radionuclides in the sediment is in the section of channel between CRM 0 and CRM 15. The longitudinal distribution in the river, based on mean sectional concentrations, is similar throughout the study reach for all radionuclides. Highest radionuclide concentrations occur at, or near, the

mouth of White Oak Creek, where the smallest sectional volume of radioactive sediment in the reach is found.

Similar patterns in the distribution of gamma radioactivity with depth in the sediment, largely controlled by the distribution of ^{137}Cs, were observed in cores collected throughout the 21-mile reach. This persistent pattern indicates that more or less regular sediment deposition, as well as net accumulation of sediment, has taken place at the sites sampled. However, there were parts of each sampling section in which either no sediment deposits were found or the sediment present was not radioactive.

The vertical distribution of ^{137}Cs in the sediment reflects annual variations in the release of this radionuclide to the Clinch River. Strong similarities in vertical distribution patterns of ^{137}Cs and ^{60}Co in the sediment suggest that incorporation of these radionuclides in Clinch River bottom sediment was due to deposition of radioactive suspended sediment originating almost exclusively from White Oak Creek.

The radioactive sediment may be classed as a clayey silt, with surprisingly little vertical variation in mineralogical composition and in cation exchange capacity. The sand- and silt-size fractions of the sediment are composed largely of quartz grains. The clay-size fraction is made up of varying mixtures of mica, clay minerals, and quartz.

INTRODUCTION

Oak Ridge National Laboratory (ORNL) began processing radioactive materials in 1943, and has released low-level liquid radioactive waste to the Clinch River via White Oak Creek since that year (see Fig. 1). This waste disposal practice has resulted in incorporation of some of the released radionuclides in bottom sediment in the river[1].

The quantity, distribution, and nature of the radioactive bottom sediment must be known in order to determine the proportion of the released radionuclides being retained in the river system, and the locus and duration of retention. With this information, the possibility of future radiation hazards resulting from current disposal practices can be assessed. An evaluation of continued use of the river for disposal of radioactive wastes can be made also. Through knowledge of the physical, mineralogical, and chemical composition of the radioactive sediment, an understanding of the mechanisms whereby the various radionuclides were incorporated in the sediment may be gained. Sampling techniques and methods of estimating and predicting radionuclide content

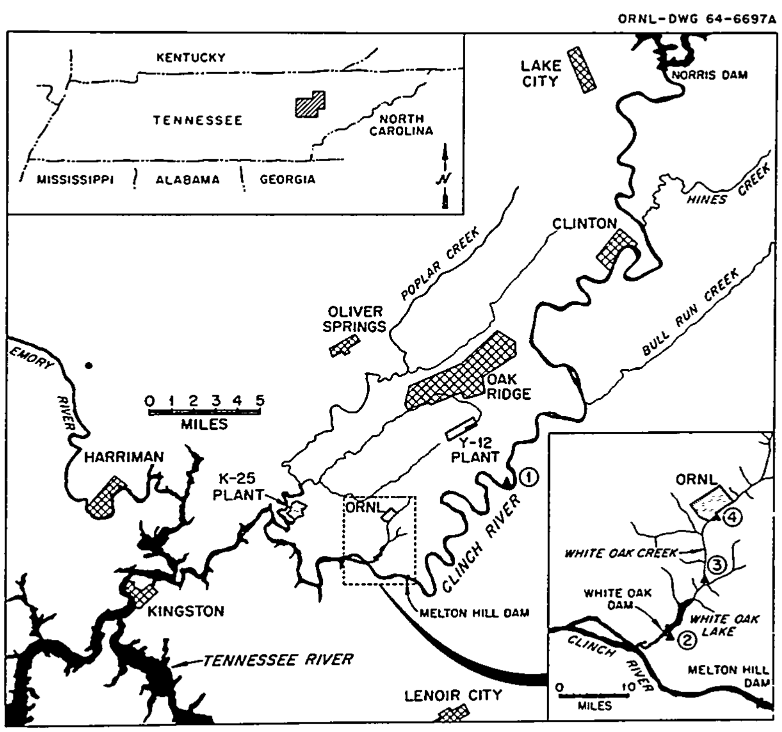

Fig. 1. Map of White Oak Creek, Clinch River, and Tennessee River in Vicinity of Oak Ridge, Tennessee. Numbers in circles indicate streamgaging station.

of the bottom sediment, developed through such a study, may suggest alternate or additional procedures that can be applied to long-term monitoring of the sediment. Bottom sediment studies thus can contribute to the realization of all five stated objectives of the Clinch River Study[2, 3]: (1) to determine the fate of radioactive materials currently being discharged to the Clinch River, (2) to determine and understand the mechanisms of dispersion of radionuclides released to the river, (3) to evaluate the direct and indirect hazards of current disposal practices in the river, (4) to evaluate the overall usefulness of this river for radioactive waste disposal purposes, and (5) to provide appropriate conclusions regarding long-term monitoring procedures.

A series of core samples of radioactive bottom sediment was taken during the summer of 1962 at 14 sampling sections in the Clinch River and at two sampling sections in each of two tributary streams, Poplar Creek and the Emory River (figure 2), by means of the Swedish Foil Sampler and other samplers. Sampling sections were selected for adequate coverage of the study reach and, wherever possible, were made to coincide with sediment ranges used by the Tennessee Valley Authority (TVA) for periodic measurements of bottom sediment accumulation or erosion. Two sampling sections were located just upstream and just downstream from the mouth of Poplar Creek to assess the effect of that tributary on distribution of radioactive sediment. The most downstream sampling section was at CRM 1.3 and the most upstream section was at CRM 22.8.

The vertical distribution of gross gamma radioactivity and the major gamma-emitting radionuclides in the cores was measured by means of a device, known as a core scanner, which was developed especially for that purpose[4]. The radioactive portions of the cores, as determined from the core scans, were mixed, sampled, and analyzed for radionuclide content in order to compute an inventory of radionuclides in bottom sediment in the Clinch River. Composite samples of 45 of the cores were analyzed for particle-size distribution.

The chemical form and associations of radionuclides in the

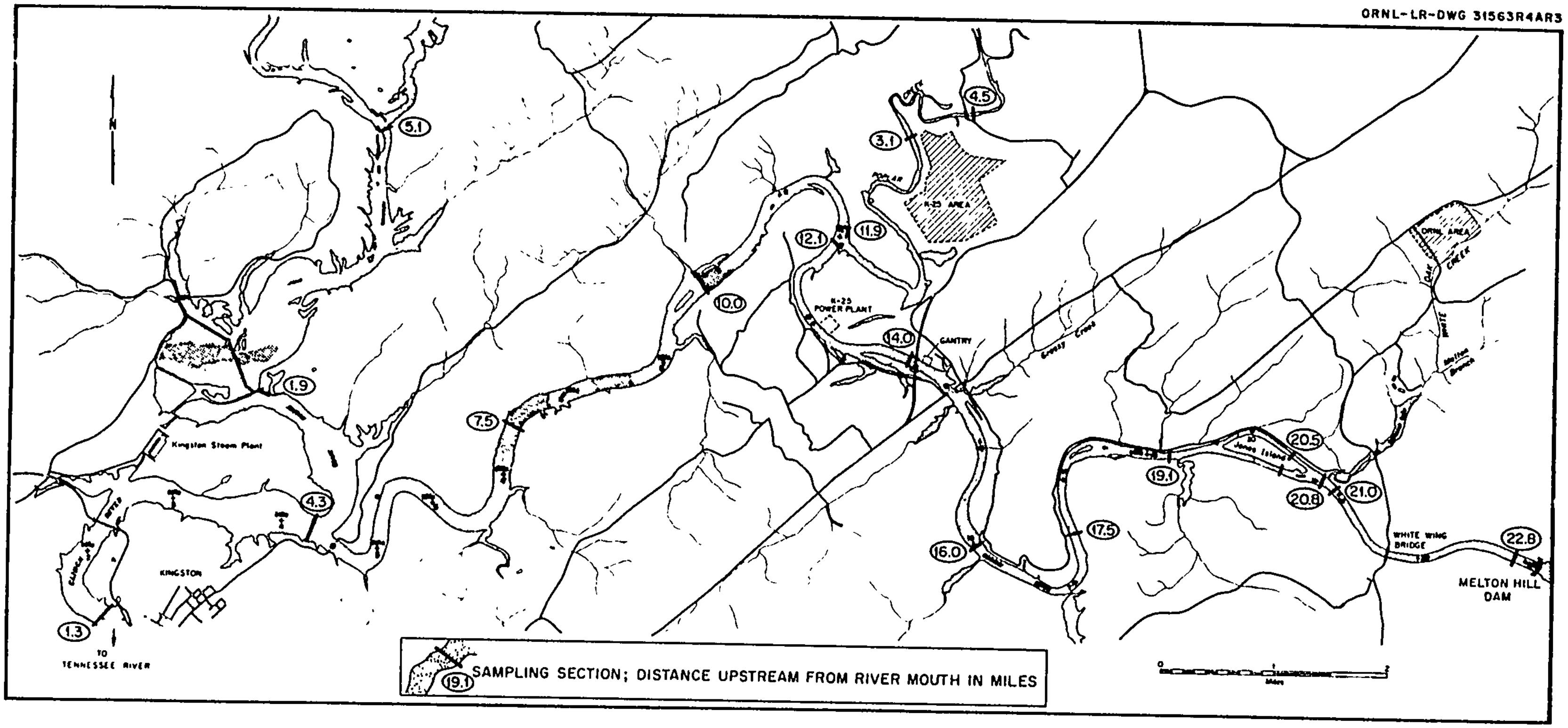

Fig. 2. Map of Clinch River Showing Locations of Bottom Sediment Core Sampling Sections[4]

bottom sediment were investigated through a program of detailed analysis of 23 sediment samples from two cores. A series of physical and chemical tests, as well as radionuclide analyses, were performed on the 23 samples in order to characterize their compositional variability. Properties measured were the mineralogy, particle-size distribution, cation exchange capacity, leachable cation content, free aluminum oxide content, free iron oxide content, inorganic and organic carbon contents and the amounts of potassium, rubidium, cesium, and strontium in each of the samples. Correlations of variations in concentration with depth for each core were determined for selected constituent pairs by means of a digital computer program in an effort to determine what properties of the sediment might be responsible for the presence of each of the radionuclides.

METHODS OF SAMPLE COLLECTION AND ANALYSES

Coring

Samplers.--Relatively undisturbed cores of bottom sediment were obtained by means of the Swedish Foil Sampler. The Swedish Foil Sampler is a 2 1/2 inch-diameter, piston-type sampler in which thin axial metal strips (foils) are used to decrease friction between the sample tube and the sediment while coring is proceeding.

Core recovery, at coring sites where radionuclides were believed to be present throughout the full thickness of the sediment penetrated, generally ranged from 80 to 100 percent. When a satisfactory core recovery was not obtained, a second coring attempt was made. At a few locations, satisfactory cores could not be obtained with the unmodified sampler. Use of a cutting shoe containing a basket-type core retainer produced adequate samples in most cases.[5]

The Swedish Foil Sampler could not be employed in 'soupy' sediments, in sandy sediments, and in sediments consisting mostly of pebbles and gravel. For 'soupy' sediments, SCUBA divers collected samples by driving thin-walled, 2 1/2-inch coring tubes into the sediment. In sandy sediments, sampling was done with a split-barrel sampler containing a basket shoe and plastic sleeve for sample retention. Hand-operated clam shell dredges were used to collect gravel samples.

Sediment cores were transferred in the field from the sample tube to plastic storage tubes, then frozen for ease of handling, to prevent slumping of soft sediment and to suppress biological and chemical action.

Selection of sampling sites.--Locations of sampling sections were based on the longitudinal pattern of radionuclide distribution in the upper portion of bottom sediment in the study reach. Sections were selected to sample reaches of high and low radioactivity, locations at which breaks in the general trend of increasing or decreasing radioactivity occurred, and reaches of transition from one radiation level to another.

Two sections established on the Emory River and on Poplar

Creek (figure 2) were for use in providing subsidiary information to health physicists at ORNL on possible movement of radioactive contaminants from the Clinch River into tributary streams.

Spacing of coring verticals within a sampling section was chosen by combined assessment of transverse distribution of sediment thickness and of radiation levels at the sediment surface. The relation of spacing of verticals to variations in thickness and radiation levels for one sampling section is shown in figure 3.

Results of coring.--Cores were collected at 135 verticals in the 14 sampling sections in the Clinch River and the sections in tributaries to the river (see figure 2). The number of verticals in a section ranged from 4 to 13.

Results of coring at each section are summarized in table 1. Two situations of special significance to the summary of coring results are emphasized by the table: (1) fifteen cores did not completely penetrate the radioactive zone; (2) not all cores or dredge samples contained significant amounts of radionuclides, as revealed by subsequent analyses (these samples were excluded from further direct consideration in computations).

Core Processing

Core processing was divided into four major procedures: (1) measurement of the distribution of gross gamma radiation in all cores and of individual radionuclides in selected cores (core counting), (2) longitudinal cutting, (3) preparation of samples for analysis, and (4) physical and radiochemical analysis of the samples. During the first two processing procedures, the cores (and dredge samples) remained frozen, as they had been since shortly after collection.

Core counting.--Variations of radioactivity with depth in the still-frozen cores were measured with a core scanner[4] (figure 4). For counting of gross gamma radioactivity, the phototube output of the core scanner was routed through a scaler and automatic timer-printout system. The number of seconds required for the accumulation of a pre-set number of counts (usually 2,048 counts) from the 2-inch increment of core opposite the collimator channel was recorded, the

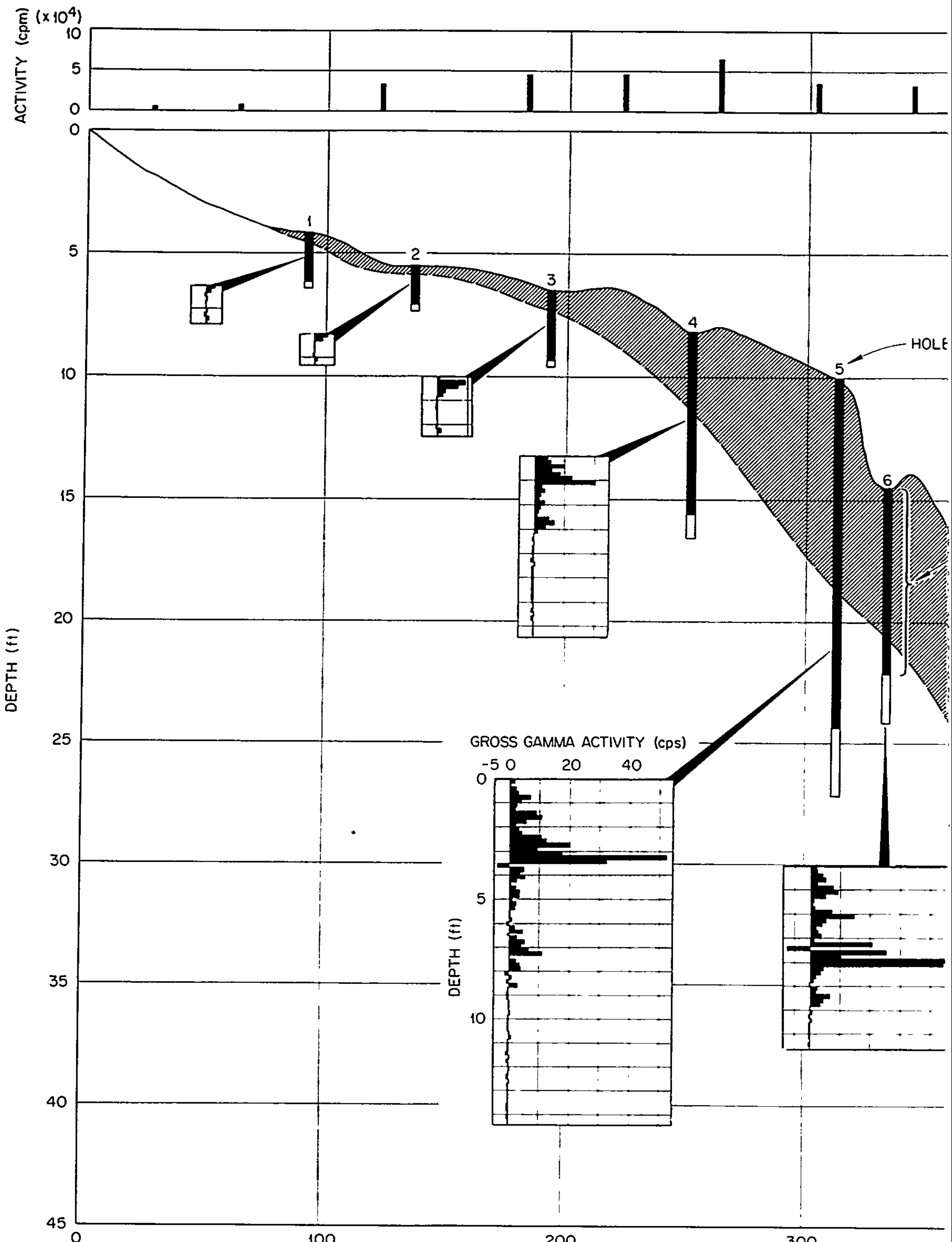

Fig. 3. Section at CRM 7.
Gamma Radioactivity Variat
Core Samples (Vertical Exa

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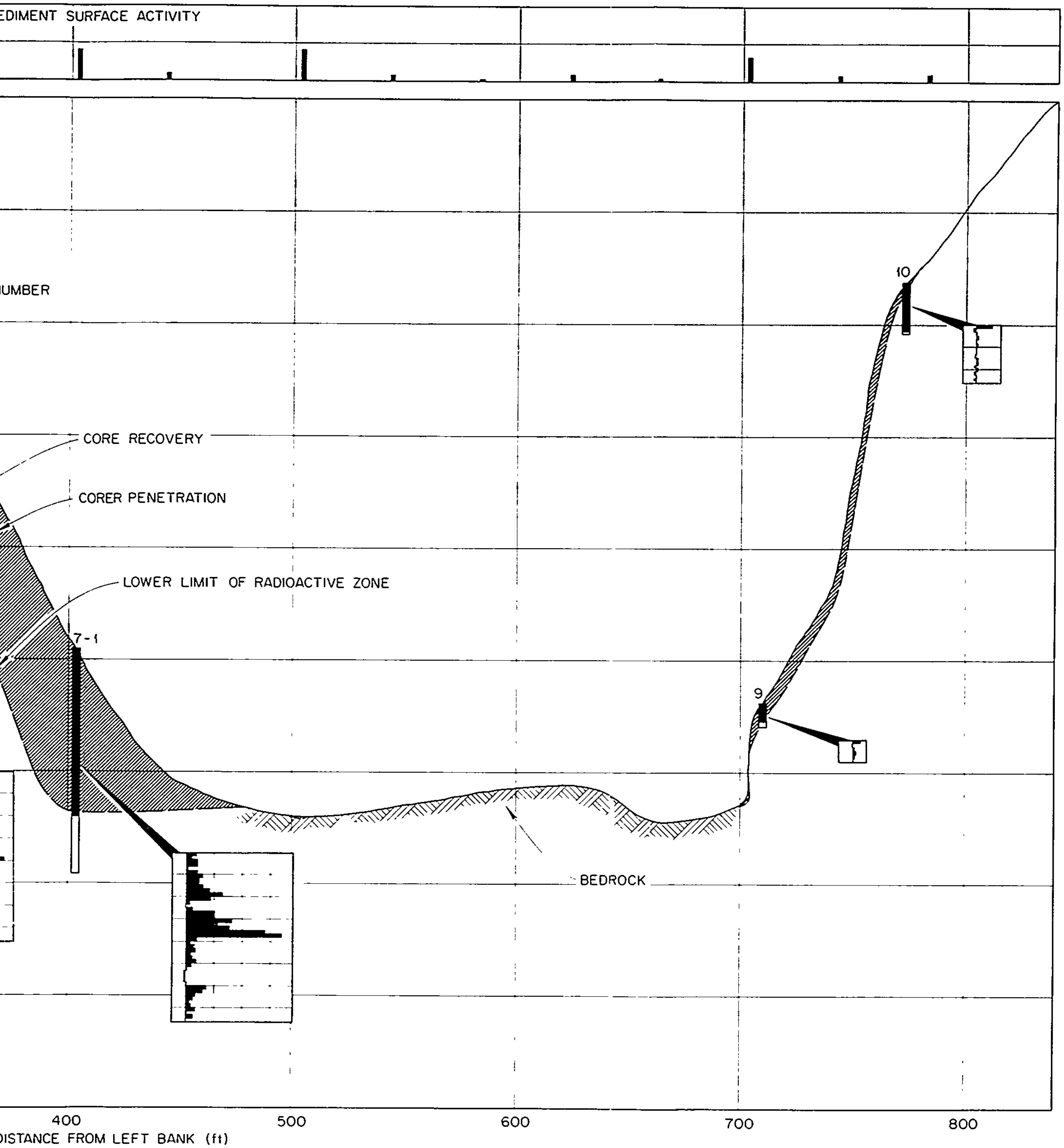

Showing Penetration, Recovery, and Gross
on with Depth for 1962 Bottom Sediment
geration 10:1)[4]

Table 1. Numbers of Sampling Sites, of Samples Used in Computations, and of Truncated Cores[a] Used in Computations for Each Sampling Section

Location of Section Miles Upstream from Mouth	Coring Verticals	Dredge Samples	Used in Computations		
			Cores	Dredge Samples	Truncated Cores
	Clinch River				
1.3	10	0	8	0	3
4.3	13	0	12	0	3
7.5	9	0	9	0	1
10.0	11	0	10	0	2
11.9	11	0	10	0	1
12.1	11	0	11	0	3
14.0	5	0	5	0	0
16.0	7	0	7	0	0
17.5	6	0	6	0	0
19.2	5	0	4	0	0
20.5	8	6	8	6	1
20.8[b]	5	7	3	7	1
21.0	4	0	2	0	0
22.8	5	1	5	1	0
	Emory River				
1.9	7	0	5	0	0
5.1	7	0	0	0	0
	Poplar Creek				
3.1	5	0	0	0	0
4.5	6	0	0	0	0

[a]Cores in which length of core recovered was less than thickness of radioactive zone.

[b]Section 100 feet downstream from mouth of White Oak Creek.

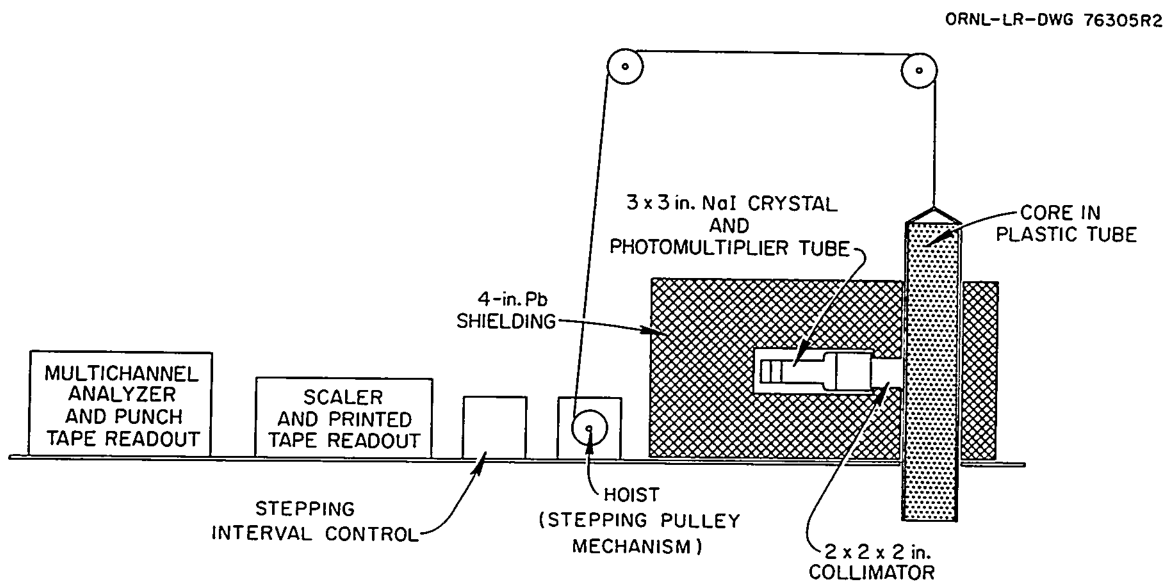

Figure 4. Diagrammatic Representation of Core Scanner[4]

core was automatically raised 2 inches by means of a calibrated hoist, and counting of the next increment of core was begun.

For gamma spectrum scanning, the phototube output of the core scanner was routed through a 512-channel pulse-height analyzer, and the analyzer output (gamma ray spectrum) for each 2-inch increment of core was recorded. The output data were then examined for analyzer malfunctions by means of a computer program. The gamma ray spectrum produced by each 2-inch increment of the cores scanned was analyzed through the use of a computer program developed at ORNL[6]. The results of each analysis, which included specific activities with computed standard deviations for eight radionuclides, were corrected for imperfect collimation and plotted for visual inspection by means of another computer program. The eight radionuclides used in the gamma spectrum analysis included the nuclear-fission products ^{137}Cs, ^{106}Ru, ^{134}Cs, $^{152, 154}Eu$, ^{144}Ce, ^{95}Zr-^{95}Nb, the neutron-activation product ^{60}Co, and the naturally-occurring radionuclide ^{40}K.

Each incremental gamma-ray radioactivity measurement was corrected for the effect of extraneous radioactivity by means of a calibration curve based on measurements of plane sources of radioactivity. The correction was applied by means of a digital computer program, which also was used for plotting the corrected incremental radioactivity against depth in the core for gross gamma scans.

Corrected counting data for each 2-inch-high slice of core were used to define the thickness of the radioactive zone at each coring vertical (see figure 3). These data were also used as a guide in determining cores in which there was no significant radioactivity.

Compositing of cores for inventory.--After core counting was completed, the cores were cut longitudinally into one half-section and two quarter-sections to provide samples for use in the inventory and for use in studies of the physicochemical characteristics of the sediments. Cutting was done by placing the frozen core in a jig and passing the jig under a carbide-tipped circular saw (figure 5).

After longitudinal sawing, one of the quarter-section cylinders of each core was cut horizontally at the base of the

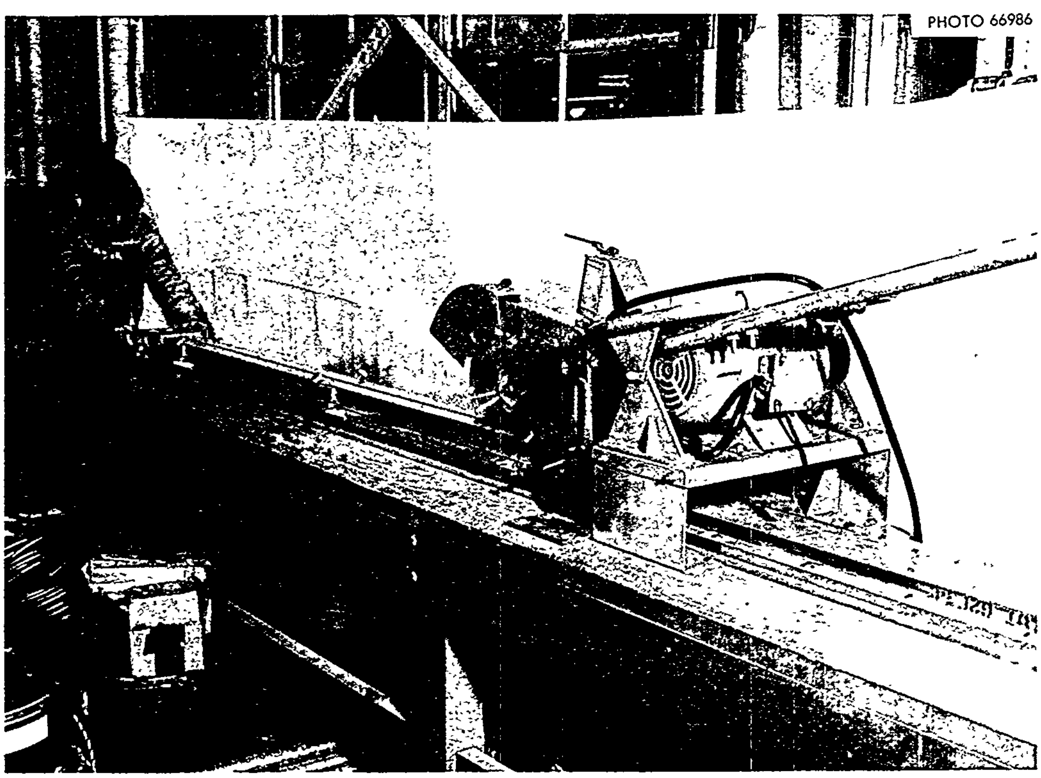

Fig. 5. Masonry Saw for Cutting Frozen Cores

radioactive zone and the non-radioactive part discarded. The radioactive part was retained for use in computing the inventory. Following cutting, the volume and weight of this cylinder of radioactive sediment were measured for determination of specific bulk weight. The volume of the cylinder (including void space) was found by permitting the frozen material to melt in a calibrated vessel. For these volumetric measurements, voids in the sediment were assumed to be completely filled with water.

A homogeneous composite sample of the radioactive quarter-sector was prepared for inventory purposes using an electrically driven egg beater. A 10-gram aliquot was split from the composite for analysis of particle size distribution. Then the remaining composite was weighed, dried at 100° C (Celsius), and reweighed, to provide data for computation of the dry-to-wet-weight ratio of the composite. A 100-gram aliquot ($\pm$ 0.01 gram) for gamma spectrum analysis, and a 20-50 gram aliquot for analyses of strontium-90 and tri-valent rare earth contents, were separated from the dry composite using a Jones soil splitter. Standard methods of chemical extraction, and beta counting of the extracts, were used in analyses[7] for ^{90}Sr and tri-valent rare earths.

Analysis of concentrations of ^{137}Cs, ^{60}Co, and ^{106}Ru in composites of the bottom sediment was made by techniques of gamma spectrometry using a 4-inch high by 4-inch diameter sodium iodide crystal for a detector and a 512-channel pulse-height analyzer. Determination of radionuclide concentrations from gamma spectra recorded on punch-tape was made by means of a computer program which utilized a technique of least-squares resolution of the gamma-ray spectra[6]. Input data for the computer program included the spectrum for the sample, the background associated with the sample, and standard spectra. Standards were prepared for ^{137}Cs, ^{60}Co, ^{106}Ru, ^{144}Ce, ^{95}Zr-^{95}Nb, ^{134}Cs, $^{152, 154}Eu$, ^{40}K and uranium and thorium ores.

To assure that results of radionuclide analyses by gamma spectrometry were reasonably accurate, nine selected samples were re-analyzed at the Low-level Counting Facility, ORNL. These samples were selected as representative of the general range of radioactivity, and of sampling location, for samples collected throughout the study reach. Analysts at this facility found that concentrations of only two radioactive contaminants, ^{137}Cs and ^{60}Co, could be reliably determined by their method of hand computation of gamma spectra data (produced from their pulse-height analyzer). Results for concentrations of ^{60}Co and ^{137}Cs determined by hand computation were slightly greater than those determined by the computer program; 7.8 and 13.8 percent, respectively. These differences are within the range, and in the direction, of error expected from such hand computation.

Physicochemical Analysis of Sediment

Sediment samples for physicochemical analysis were taken from two cores which had radioactive zones several feet thick. The samples consisted of altérnate 2-inch slices of sediment from the two cores. Comprehensive particle-size analyses of aliquots of the 23 samples were made by wet seiving and bottom withdrawal tube[8]. The method of size analysis used involves dispersal of the sediment particles by chemical means, which can be expected to break up aggregates of clay minerals which may have existed in the bottom sediment as silt- or sand-size masses. The method thus results in a determined particle-size distribution which is not necessarily fully representative of the actual distribution of particle sizes in the natural environment. However, it gives a measure of the ultimate size distribution, which is useful in hydraulic studies of sediment movement and in mineralogical studies of sediment composition.

Particle-size separations of aliquots of the same samples into sand, silt, and clay were performed by wet seiving and centrifugation after the samples had been initially dried. Dispersal was by hand stirring or, for some samples, by ultrasonic agitation. This method of separation tended to preserve aggregates of smaller particles, but additional aggregates may have formed as a result of the attachment of clay-size particles to sand- and silt-size particles when the samples were dried. Leaf fragments and twigs tended to be concentrated in the $<$ 2 micron clay-size fraction following centrifugation. Determinations of cation exchange capacities and mineralogical composition were made of the three size fractions.

Cation exchange capacities were determined by means of an ammonium chloride leach followed by measurement of the sorbed ammonium by distillation. The content of leachable cations in the raw sediment samples was determined through a chemical analysis of the leachates obtained as a result of the cation exchange capacity determinations. Mineralogical determinations were made by X-ray diffractometer.

The content of free alumina in the samples was determined by extraction in a 0.5 N solution of hydroxide[9] and colorimetric analysis by the "aluminon" method[10]. Free iron oxides were extracted in a buffered dithionate-citrate solution[11] and analyzed by the potassium thiocyanate method[10]. The extraction methods used are believed to have caused very little dissolution of silicate minerals present in the sediment.

The content of organic carbon in each of the sediment samples was determined as the difference between the total amount of carbon dioxide evolved through combustion of the sample at 1350° C and the carbonate-derived carbon dioxide as determined by a low-temperature gas-evolution technique[12]. Analysis of the sample for minor and trace elements was performed by flame spectrophotometric methods, using recently-developed precision methods for the determination of strontium[13] and cesium[14]. The content of the radionuclides in each sample was determined by the same method as that used for analysis of composite samples.

Method of Computing Inventory

The volume of radioactive sediments in the Clinch River was divided into subvolumes in computing the inventory. The quantity of a radionuclide in a subvolume is equal to the product of its concentration (curies per unit weight) and weight of sediment in the subvolume. The total quantity of the radionuclide in the study reach is the sum of its quantities in all the subvolumes.

Locations of sampling sections and of coring verticals in the study reach served as controls for dividing the volume of sediment into subvolumes. Each subvolume is associated with a specific coring vertical.

Each subvolume is assumed to be prismatic in shape; its length extends from midpoint to midpoint between adjacent sampling sections; its width extends from midpoint to midpoint between adjacent coring verticals in the sampling section, and its depth is equal to the thickness of the radioactive zone for the specified core. These definitions of dimensions of a subvolume are modified, first, for the two terminal subreaches and, second, for terminal verticals (where thickness of sediment is zero) in a sampling section.

The widths of radioactive zones in the sampling sections did not in every case extend from one edge of water to the other. Hence, the transverse locations of at least two, and sometimes more, terminal verticals of radioactive zone(s) in a section had to be defined. These terminal verticals were defined on the basis of (1) locations of cores that contained no detectable radioactivity, (2) rates of change of the gamma radiation levels at the surface of the bottom sediments, (3) rates of change of thickness of the radioactive zone, (4) transverse slope of the stream bed, and (5) limits of bedrock areas as determined by probing of the bed material. The influence of some of these factors on location of terminal verticals is indicated in figure 3.

The weight of sediment in a subvolume is the product of its volume and the specific bulk weight and dry-to-wet weight ratio of its associated core. Specific bulk weight is the wet weight per unit volume. Weight of the subvolume was converted to an oven-dry basis by the dry-to-wet weight ratio because oven-dried (100° C) composites were used in radiochemical analyses.

Approximately two years elapsed between collection and radiochemical analyses of cores. During this period decrease in radionuclide content through the process of radioactive decay was appreciable for some radionuclides. Concentrations used in the computations were adjusted to those at the time of sampling except for rare earths. No adjustment for radioactive decay of rare earths was made because they are a mixture of radionuclides having a variety of radioactive half-lives. Methods of adjustment for effects of radioactive decay on radiation have been described by the U. S. Public Health Service[15].

Computation of the inventory was facilitated through use of a general purpose digital computer. Specific bulk weight, dry-to-wet weight ratio, and area were computed and checked manually prior to use as input data in computer programs.

Thicknesses of radioactive sediment for truncated cores and dredge samples were estimated as follows:

Distribution patterns of gross gamma radiation were discernable in many truncated cores. Patterns of radioactivity distribution in such cores were compared, using correlation analyses (see gross gamma core scans), to patterns for cores which penetrated the entire radioactive zone at nearby coring verticals. From results of these comparisons, the depth to which radioactivity extended could be estimated for truncated cores.

In some instances the truncated length was too short to clearly discern the distribution pattern of radioactivity, in which case the radioactive thickness at the coring vertical was assumed to be equal to the depth of penetration of the coring tool. Fortunately, the fraction of core recovered was large in most cases.

Depth of penetration of hand-operated dredges could not be measured. Depths were computed using information on volume of sample collected and on geometric characteristics of the sampler. Computed depth of sampling was assumed to be the radioactive depth.

INVENTORY

In the course of acquiring necessary data to inventory radioactivity in Clinch River bottom sediment, information has been gathered not only on quantities of radionuclides in the sediment, but also on the distribution of radionuclides in these sediments and the distribution of the radioactive sediments in the river.

Inventory of Identified Radionuclides

On the basis of radionuclide analyses of composite samples of bottom sediment cores from the Clinch River, the following quantities of radionuclides are associated with bottom sediments of the river between CRM 0.0 and CRM 21.0; 150 curies of ^{137}Cs, 18 curies of ^{60}Co, 16 curies of ^{106}Ru, at least 10 curies of rare earths, and 2.9 curies of ^{90}Sr.

All quantities have been adjusted for effects of radioactive decay to the quantities present at the time of sampling (July 1, 1962) except for the rare earths (see Method of Computing Inventory). The quantity of rare earths reported is that present at the time of radiochemical analysis (June 1964).

? 5% 37CS

Total inventoried and identified radioactivity in the river is 200 curies, of which ^{137}Cs constitutes 77 percent; ^{60}Co, 8.7 percent; ^{106}Ru, 7.7 percent; rare earths, 5.1 percent; and ^{90}Sr, 1.5 percent.

About 95 percent of the identified radioactivity is in the reach of river downstream from CRM 15 (table 2). At least 50 percent of the identified radioactivity is downstream from CRM 8.7.

Estimate of accuracy of inventory.--The quantity of a radionuclide in a volume of sediment, R, may be expressed by the equation

$$R = C \times b\ t\ \gamma_s \frac{w_d}{w_w} ,$$

in which the independent variables are, respectively, concentration of a radionuclide, length of subreach, width of sampling section, mean thickness of radioactive sediment in sampling section, specific bulk weight of the sediment, and dry-to-wet weight ratio. The

Table 2. Total Identified Radioactivity and Volume of Radioactive Sediment in Subreaches

Location of Subreach		Total Identified Radioactivity, in Curies		Volume in Acre-Feet	
Begin (CRM)	End (CRM)	In Subreach	Cumulative	In Subreach	Cumulative
0	2.80	22	22	340	340
2.80	5.90	42	64	380	720
5.90	8.95	54	118	480	1,200
8.95	10.95	46	164	430	1,630
10.95	12.00	9.3	173	93	1,720
12.00	13.05	10	183	85	1,810
13.05	15.00	6.8	191	38	1,850
15.00	16.75	2.2	193	33	1,880
16.75	18.35	4.7	198	39	1,920
18.35	19.85	.1	198	4.7	1,920
19.85	20.65	2.3	200	5.9	1,930
20.65	20.90	.1	200	1.4	1,930
20.90	21.00	.2	200	.3	1,930

following expression is used to compute the level of expected error in R:

$$\sigma_R = R \sum_{1}^{6} \frac{\sigma_z}{Z} ,$$

in which σ_R is the standard deviation for R, σ_z is the standard deviation for each independent variable Z enumerated above, and $\frac{\sigma_z}{Z}$ is the relative error or coefficient of variation, for Z.

The relative error in the measurement of physical properties which appear in the above equation has been estimated by comparing the precision in measuring each property to the magnitude of the property. Errors in measuring specific bulk weight and dry-to-wet weight ratio are less than 5 and 2 percent, respectively. Relative errors in dimensions of sediment volume have been estimated for each subreach downstream from CRM 15. Limitation in considered reach length has been made because 95 percent of the radioactivity is downstream from CRM 15. Coefficients of variation for subreach length, width, and thickness are about 3, 5, and 15 percent, respectively. Other independent sources of error in the determination of radionuclide concentration are accuracy of radionuclide standards, counting errors in analysis, and sampling errors. Error in radionuclide content of standards is about 5 percent. Average levels of counting errors for cesium-137, cobalt-60, ruthenium-106, strontium-90, and rare earths are 2, 8, 23, 4, and 2 percent, respectively.

The estimated errors in the inventory (neglecting sampling error and using the foregoing expression for σ_R) are as follows: 18 percent for ^{137}Cs, ^{90}Sr, and rare earths; 20 percent for ^{60}Co; and 29 percent for ^{106}Ru.

Retention Factors

The retention factor for a radionuclide is defined as the ratio, expressed in percent, of the quantity of the radionuclide in the sediment in July 1962 to that quantity released to the river during the period December 1943 to July 1962.

The magnitude of the retention factor is independent of time if radioactive decay is taken into account.

$$k_r = \frac{I - O}{I}(100) = \frac{R}{I\,e^{-\lambda T}}(100) ,$$

$$I = \int_0^T i\,dt, \text{ and } O = \int_0^T o\,dt ,$$

in which k_r = retention factor, in percent,

I = inflow of radionuclide to study reach during the time period T, in curies,

O = outflow of radionuclide from study reach during time period T, in curies,

R = quantity of radionuclide in bottom sediment of study reach at end of period T, in curies,

λ = radioactive decay constant for radionuclide, in sec^{-1},

T = duration of time period, in seconds,

i = rate of inflow of radionuclide to study reach, in curies per second, a function of time,

o = rate of outflow of radionuclide from study reach, in curies per second, a function of time, and

t = time, in seconds.

R may be expressed as a function of inflow and outflow if the effect of radioactive decay is considered.

$$R = \int_0^T i e^{-\lambda(T - t)}\,dt - \int_0^T o e^{-\lambda(T - t)}\,dt$$

$$= e^{-\lambda T}\int_0^T i\,dt - e^{-\lambda T}\int_0^T o\,dt$$

$$= e^{-\lambda T}(I - O)$$

Hence, $k_r = \frac{R}{I\ e^{-\lambda T}}\ (100) = \frac{e^{-\lambda T}(I - 0)}{e^{-\lambda T}\ I}\ (100) = \frac{I - 0}{I}\ (100)$

and is independent of time.

Only annual records of inflow are available so it is necessary to use numerical techniques to compute k_r:

$$k_r = \frac{R_m/e^{-\lambda T_2}}{\sum_{n=1}^{T} i_n e^{-\lambda(T-n)}} \quad (100), \quad n = 1, 2, \ldots\ldots, T \text{ years} \qquad (1)$$

in which $R_m = R\ e^{-\lambda T_2}$ = inventory of radionuclide at time of radiochemical analysis, in curies,

λ = radioactive decay constant, in years^{-1},

T_2 = time between collection of cores and radiochemical analysis, in years,

i_n = annual inflow of radionuclide to study reach during n^{th} year, in curies per year.

Using equation 1 the retention factors are computed to be 21 percent for ^{137}Cs, 9 percent for ^{60}Co, 0.4 percent for ^{106}Ru, and 0.2 percent for ^{90}Sr. Retention of the rare earths may approach, and possibly exceeds, 25 percent (see Ref. 16). A good estimate of the retention factor for rare earths has not been obtained because mixtures of radioelements of undetermined composition are involved. The above estimate was derived from a conservative consideration of the ^{144}Ce and other rare earths contents of three samples.

Contributions to Inventory From Reaches Outside of Study Reach

Contributions to the radionuclide content of bottom sediments in the study reach from fallout from weapons tests appear to be negligible. Concentrations of radionuclides in bottom sediment at sampling section 2.0 miles upstream from the study reach, CRM 22.8, are below limits of detection. Concentrations of ^{137}Cs and ^{60}Co in waters flowing past a water-sampling station at CRM 41.5, upstream from the study reach, have been at or below limits of detection since the study was begun. No rare earths have been detected in water samples collected at this station. Small concentrations of ^{106}Ru and ^{90}Sr have been detected regularly in water samples,[17] but these radionuclides constitute a minor fraction of the total inventory, and their retention factors are low.

Cores were collected in Poplar Creek and Emory River to gage the extent of upstream movement of contaminated waters from the Clinch River. Hydraulic conditions are such in the study reach of the Clinch River that upstream movement of water from the river can occur in any tributary.

Examination of gross gamma radioactivity scans of cores from Poplar Creek and from the section at mile 5.1 on the Emory River indicate that the radionuclide content in bottom sediments at these sampling sites (Fig. 2) is negligible. However, concentrations of radioactivity in some cores from mile 1.9 on the Emory River were above lower limits of detection. Furthermore, Pickering, in a study of radioactivity in bottom sediments of sloughs adjacent to the Clinch River[4], noted that radiation levels in some areas approached levels measured at the surface of Clinch River bottom sediment. This evidence of radioactivity in bottom sediments in the lowermost reaches of tributaries to the Clinch River suggests that supplemental inventories in these areas might be advisable.

DISTRIBUTION OF RADIOACTIVE SEDIMENT

Variation of Radioactivity With Depth in Cores

Gross gamma core scans.--Scanning the bottom sediment cores for variations in gross gamma radioactivity[1/] with depth revealed the general pattern of variation in each core and provided a basis for determining the thickness of radioactive sediment at each sample site. The results of the core scanning indicated that the entire thickness of radioactive sediment had been sampled in most cores. The core showing the greatest thickness of radioactive sediment, 8.7 feet, was obtained at CRM 7.5.

The results of gross gamma scanning of core samples from CRM 7.5 are shown in Figure 3. Sediment deposition, and therefore accumulation of radioactive sediment, has been largely confined to the more gently-sloping left half of the stream channel which constituted the side of the former stream channel and a portion of the flood plain of the stream prior to inundation by Watts Bar Lake. (Note that the vertical exaggeration of the cross section plot is 10:1). Portions of the cross section showing a water depth of less than 6 feet are not submerged during low winter lake levels. The resulting non-deposition of sediments and erosion probably account for the very thin layer of radioactive sediment found in cores from such sites (cores 1 and 2).

Cores 4, 5, 6, and 7-1 in Figure 3 show a persistent (general) pattern of variation of gross gamma radioactivity with depth. Similar distribution patterns of gross gamma radioactivity were observed in several cores from other sampling sections in the portion of the study reach downstream from CRM 18. The general pattern which they exhibit is strikingly similar to the pattern of annual releases of ^{137}Cs to

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[1/] As used in this report, "gross gamma radioactivity" refers to the measured quantity of radioactivity given off by all gamma ray-emitting nuclides in the sample. The fraction of the total radioactivity detected depends on the design and sensitivity of the analytical equipment used.

the Clinch River as measured at White Oak Dam (Table 3). This similarity is illustrated in Figure 6 for several of the cores. The zone of highest radioactivity in each of the four cores is assumed to correspond to the period during which releases of ^{137}Cs were highest -- i.e., 1956[2].

The patterns of variation of gross gamma radioactivity with depth in the sediment cores were compared by means of a digital computer program. With this program, the variation of radioactivity in a core of any length could be compared to that in any other core, provided two reference points at different depths could be assigned as corresponding to the same parts of the patterns in each of the cores compared. Core lengths were adjusted for the comparison on the basis of the distance between the two reference points picked for each core. A series of correlations were attempted for each pair of cores, using a different adjustment factor in each correlation attempt. The output of the program included the coefficients of correlation and of regression for the best correlation attempted with each pair of cores.

Coefficients of correlation resulting from a comparison of the four cores shown in Figure 6 to annual releases of ^{137}Cs to the Clinch River are included in Table 4.

By means of the computer program, gross gamma scans of 27 cores which exhibited a similar pattern of variation of gross gamma radioactivity with depth were compared to the pattern of annual releases of ^{137}Cs to the Clinch River, corrected for decay to the date of sampling. Coefficients of correlation ranged from 0.60 to 0.90 and had a mean of 0.76. When annual releases of ^{60}Co were added to those of ^{137}Cs, and correction made for the relative gamma ray emission of the two nuclides, the comparison showed improved correlation with the cores, giving a mean coefficient of correlation of 0.80 and a range of 0.64 to 0.98. When the annual releases of ^{106}Ru were added to those of ^{137}Cs and ^{60}Co, after making corrections for relative gamma ray emission, the correlation was not as good, giving a mean coefficient or correlation of 0.68 and a range of 0.44 to 0.97.

The correlation results suggest that the pattern of variation

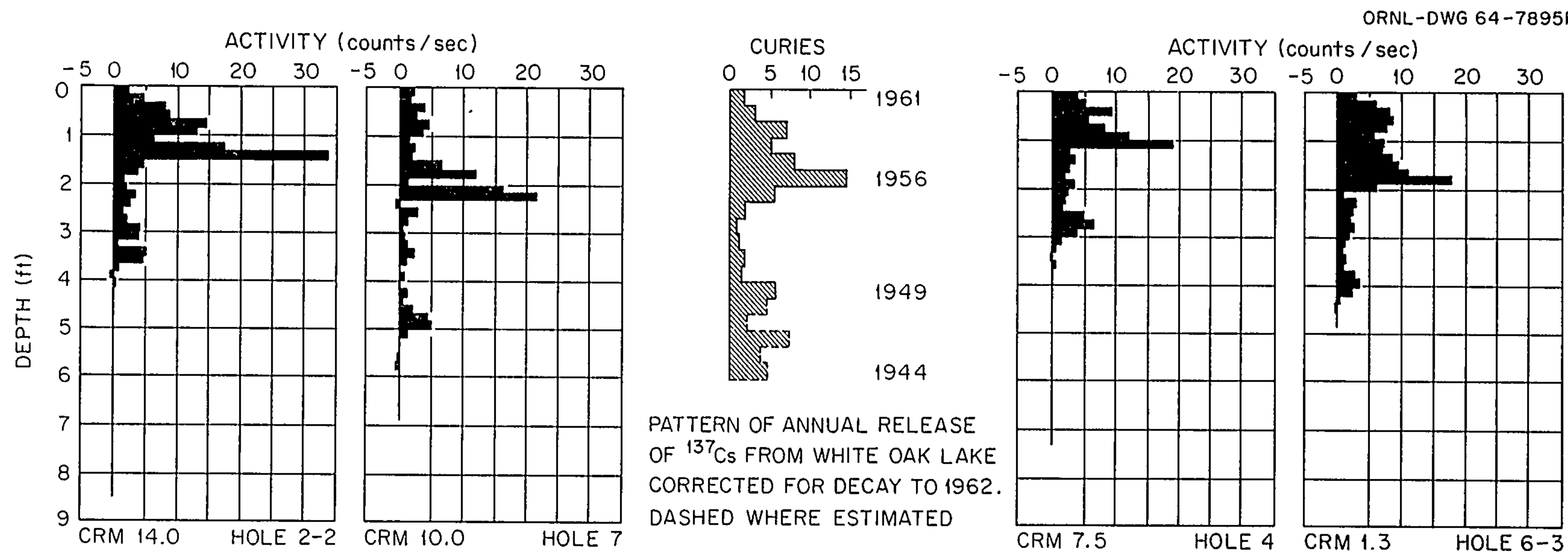

Fig. 6. Comparison of Patterns of Variation with Depth of Gross Gamma Radioactivity in Four Bottom Sediment Cores to Variations in Annual Releases of ^{137}Cs to Clinch River

Table 3. Yearly Discharges of Radionuclides to the Clinch River (curies)[1]

Year	Gross Beta	^{137}Cs	^{106}Ru	^{90}Sr	TRE(-Ce)	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co
1949	718	77	110	150	77	18	180	22	77	
1950	191	19	23	38	30		15	42	19	
1951	101	20	18	29	11		4.5	2.2	18	
1952	214	9.9	15	72	26	23	19	18	20	
1953	304	6.4	26	130	110	6.7	7.6	3.6	2.1	
1954	384	22	11	140	160	24	14	9.2	3.5	
1955	437	63	31	93	150	85	5.2	5.7	7.0	6.6
1956	582	170	29	100	140	59	12	15	3.5	46
1957	397	89	60	83	110	13	23	7.1	1.2	4.8
1958	544	55	42	150	240	30	6.0	6.0	8.2	8.7
1959	937	76	520	60	94	48	27	30	0.5	77
1960	2190	31	1900	28	48	27	38	45	5.3	72
1961	2230	15	2000	22	24	4.2	20	70	3.7	31
1962	1440	5.6	1400	9.4	11	1.2	2.2	7.7	0.36	14
1963	470	3.5	430	7.8	9.4	1.5	0.34	0.71	0.44	14

[1] Calendar year basis, after Cowser and Snyder[30]

Table 4. Coefficients of Correlation for Comparison of Vertical Distribution of Gross Gamma Radioactivity in Sediment Cores with Annual Releases of Radionuclides[a]

Sampling Section (CRM)	Core Number	^{137}Cs[a]	$^{137}Cs + ^{60}Co$[a]	$^{137}Cs + ^{60}Co + ^{106}Ru$
1.3	3	0.64	0.67	0.68
1.3	5	0.78	0.84	0.72
1.3	6-2	0.63	0.72	0.64
1.3	6-3	0.73	0.78	0.78
1.3	7	0.60	0.64	0.65
1.3	7-2	0.88	0.93	0.80
4.3	2-2	0.67	0.83	0.69
4.3	11-2	0.73	0.79	0.63
7.5	4	0.81	0.82	0.62
7.5	5	0.80	0.72	0.51
7.5	6-1	0.81	0.75	0.49
7.5	7-1	0.75	0.79	0.57
7.5	7-2	0.72	0.64	0.50
10.0	2	0.83	0.82	0.61
10.0	3-3	0.77	0.90	0.81
10.0	7	0.83	0.79	0.44
11.9	9	0.76	0.76	----[b]
11.9	10	0.60	0.79	0.71
12.1	2	0.88	0.89	0.80
12.1	6-1	0.66	0.82	0.80
12.1	7	0.79	0.78	0.77
12.1	9	0.89	0.98	0.87
14.0	2-1	0.82	0.85	0.60
14.0	2-2	0.80	0.84	0.62
14.0	3	0.76	0.87	0.97
17.5	3-1	0.90	0.79	0.83
17.5	3-2	0.76	0.66	0.63
Mean		0.76	0.80	0.68
Standard Deviation		0.084	0.083	0.126

[a]Corrected for decay to date of sampling. Where releases are combined, correction has been made for the relative gamma emission of the radionuclides.

[b]Dashes indicate correlation not obtained.

in gross gamma radioactivity exhibited by the cores tested reflects not only the pattern of annual releases of ^{137}Cs to the Clinch River, but to a minor extent, that of annual releases of ^{60}Co as well.

Gamma spectrum core scans.--Nine cores were selected for gamma ray spectrum scanning on the basis of gross gamma scan results. Cores selected were those with a zone of radioactive sediment which was several feet thick and which showed the characteristic pattern of variation in gross gamma radioactivity with depth illustrated in Figures 3 and 6.

Four gamma-emitting radionuclides were present in the bottom sediment in concentrations high enough to provide acceptable data for the full thickness of the radioactive zone in each core scanned; they were ^{137}Cs, ^{60}Co, ^{106}Ru, and ^{40}K. Because of its short half-life (1 year), ^{106}Ru was detected in only the upper portions of most of the cores. The presence of the naturally-occurring radionuclide ^{40}K was not due to releases from ORNL.

Variations in concentrations with depth of the three most abundant gamma-emitting radionuclides in a core from Hole 6, CRM 7.5 are illustrated in Figure 7 (compare with Fig. 3): values whose standard deviations represented greater than 75 percent of the analytical values computed prior to collimation correction, have been plotted as zero values.

An examination of the first four plots in Figure 7 demonstrates the relative contributions of the three radionuclides to the total gamma ray radioactivity of the core. Over 80 percent of the radioactivity is contributed by ^{137}Cs. The contribution of radioactivity from ^{106}Ru is confined to the upper portion of the core. (The isolated positive values for ^{106}Ru below a depth of 20 inches on the plot are questionable). Similar results obtained for the eight other cores that were gamma spectrum scanned are shown in Table 5. These results compare favorably with results of the inventory of radionuclides in bottom sediments in the Clinch River (see Inventory).

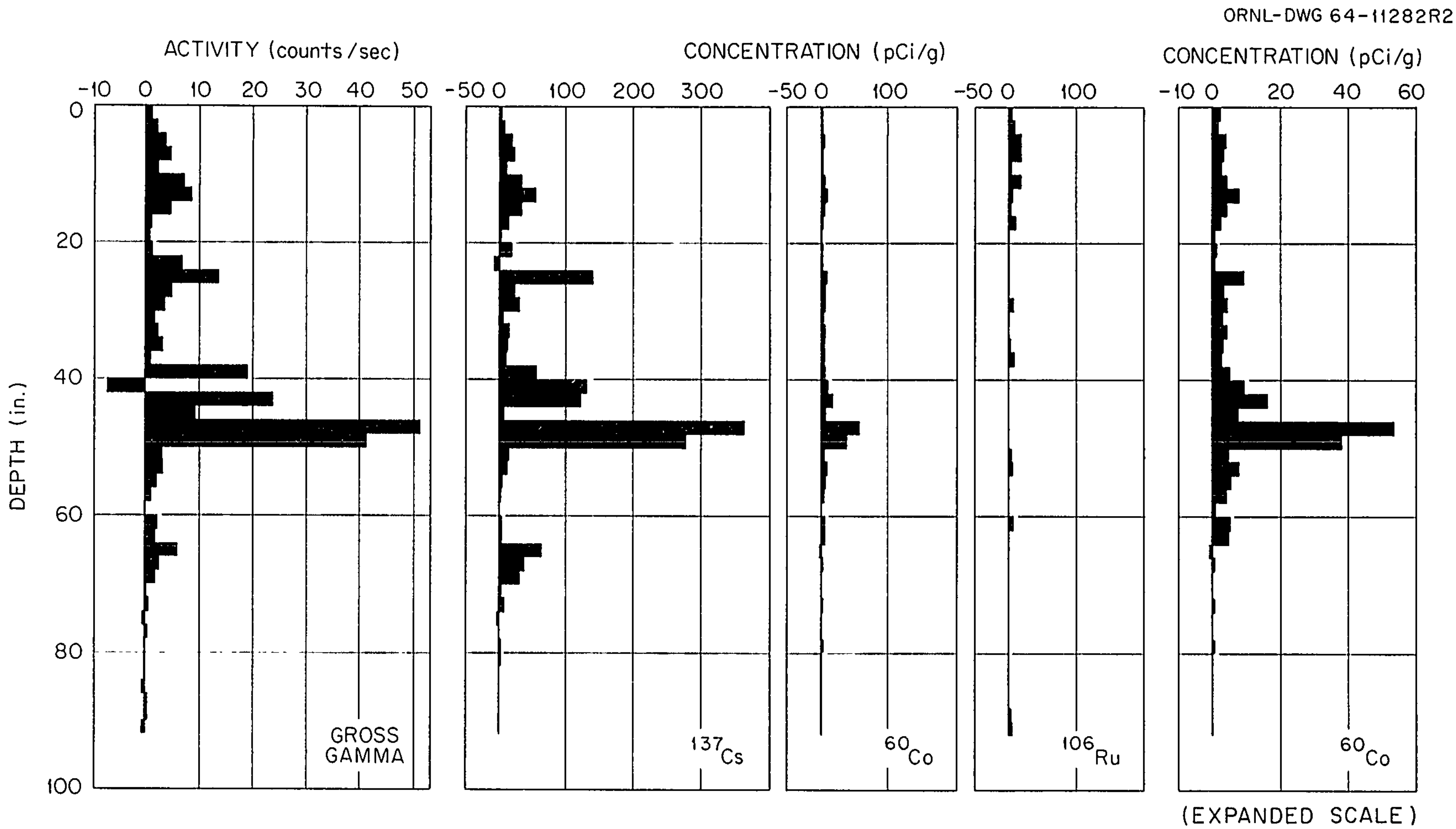

Fig. 7. Variations with Depth in Concentrations of ^{137}Cs, ^{60}Co, and ^{106}Ru in Hole 6, CRM 7.5

Table 5. Fractional Contributions of ^{137}Cs, ^{60}Co, and ^{106}Ru to Total Gamma-Ray Radioactivity of Selected Clinch River Sediment Cores

CRM	Hole Number	^{137}Cs Fraction	^{60}Co Fraction	^{106}Ru Fraction
1.3	3	0.82	0.11	0.07
1.3	6-3	0.77	0.13	0.10
7.5	5	0.81	0.13	0.16
7.5	6	0.82	0.12	0.06
7.5	7-2	0.77	0.13	0.10
10.0	2	0.79	0.10	0.11
14.0	2	0.80	0.13	0.07
14.0	2-2	0.84	0.11	0.05
17.5	3-1	0.86	0.10	0.04
Mean Value		0.81	0.12	0.07

The similarity between the distributions of ^{137}Cs and ^{60}Co with depth in the core can be observed with the aid of the fifth plot in Figure 7, which shows variations in ^{60}Co concentration with depth at an expanded scale. The same relationship was exhibited by the eight other cores gross gamma scanned. Statistical treatment of the relationship between ^{137}Cs and ^{60}Co in all nine cores further emphasizes the similarity in distribution of the two radionuclides (see Table 6).

Relationship of Vertical Distribution to Annual Releases

Results of radiochemical analyses of water samples collected weekly at White Oak Dam for a period of 2 years indicate that on the average nearly 70 percent of the ^{137}Cs present in the water is associated with suspended solids having a diameter greater than 0.7 micron.[17] Cesium is known to be preferentially sorbed by layered aluminosilicate minerals in which the c-axis spacing is 10 angstroms.[18, 19] Such minerals are abundant in White Oak Creek basin.[20, 21] Furthermore,

Table 6. Relationship of ^{60}Co to ^{137}Cs in Selected Bottom Sediment Cores From the Clinch River

CRM	Hole Number	Correlation Coefficient	Regression Coefficient	Ratio of Mean Concentrations (Cs/Co)
1.3	3	0.62	0.09	7.39
1.3	6-3	0.89	0.12	5.83
7.5	5	0.90	0.13	5.77
7.5	6	0.93	0.13	7.34
7.5	7-2	0.91	0.13	5.86
10.0	2	0.87	0.11	7.83
14.0	2	0.90	0.13	6.37
14.0	2-2	0.87	0.13	7.25
17.5	3-1	0.99	0.09	9.11
Mean Value		0.88	0.12	6.97

this sorption reaction is time dependent and only slowly reversible. It is safe to assume, then, that incorporation of ^{137}Cs in Clinch River bottom sediment is primarily a result of sedimentation of suspended radioactive aluminosilicate minerals which enter the river in water from White Oak Creek.

The year of highest release of ^{137}Cs to the Clinch River was 1956. This high release resulted from the draining of White Oak Lake during the latter half of 1955[2], and exposure to erosion of the accumulation of fine sediment on the lake bottom. Rapid erosion of the exposed radioactive lake sediment during the subsequent period of high winter rainfall resulted in an increase in the quantity of radioactive sediment entering the Clinch River, and higher content of ^{137}Cs in bottom sediment deposited in the river during 1956. Through the operation of processes such as this, variations in annual releases of ^{137}Cs have been recorded as variations in the vertical distribution of ^{137}Cs in the bottom sediment in portions of the river where more or less regular and persistent deposition of sediment has taken place.

The pattern of variation in the content of ^{60}Co in the sediment does not strongly resemble the pattern of annual releases of ^{60}Co to the Clinch River (Table 3). Some effect of annual releases is implied, however, by the improved correlation which resulted when annual ^{60}Co releases were taken into account in the comparison of gross gamma core scan patterns described previously. The most striking feature of the pattern of ^{60}Co variation in the cores gamma spectrum scanned is its great similarity to the pattern of ^{137}Cs variations (see Fig. 7 and Table 6). This similarity suggests that ^{60}Co, like ^{137}Cs, was incorporated in Clinch River bottom sediment primarily by sedimentation of suspended solids which had obtained their content of ^{60}Co before entering the Clinch River. It also suggests that a rather small fraction of the total ^{60}Co released to the river became incorporated in Clinch River bottom sediment; otherwise, distribution of ^{60}Co in the cores would resemble the pattern of annual releases of ^{60}Co to the river. This second suggestion is supported by the results of water sampling which show that only 20 percent of the ^{60}Co released from White Oak Creek is associated with suspended sediment with a diameter greater than 0.7 micron.[17] It is confirmed by the results of the inventory of radionuclides in the bottom sediment (see Retention Factors). Although the results of water sampling do not preclude association of the remaining 80 percent of the ^{60}Co with smaller-diameter suspended solids, some of which might be deposited in the Clinch River, it is possible that the nuclide may be present in White Oak Creek water also in a second form, such as a dissolved ion. The presence of two chemical forms of ^{60}Co in White Oak Creek water does not conflict with the results of gamma spectrum core scanning if it is assumed that the ratio of dissolved ^{60}Co to solids-associated ^{60}Co in the water did not remain constant during the period in which the sampled sediment was deposited.

The suggestion that some of the ^{60}Co, like most of the ^{137}Cs, is associated with suspended solids when it enters the Clinch River from White Oak Creek does not necessarily imply that the two radionuclides are associated with the same type of solid. It is assumed that ^{137}Cs is primarily associated with layered aluminosilicate minerals. Leaching experiments[4] have indicated that ^{60}Co may be present in the bottom sediment as a hydrated oxide. The studies further indicate that ^{60}Co, like ^{137}Cs and ^{106}Ru, is not readily removed by leaching after it becomes associated with the bottom sediment. Other studies have indicated that ^{60}Co in the sediment is associated with iron and manganese oxides.[22]

The contribution of ^{106}Ru to gamma radioactivity in the upper portion of the sediment cores studied is much less than one would expect if its relative retention in the sediment were as great as that of ^{137}Cs. It can be assumed, then, that a relatively small fraction of the total amount of ^{106}Ru that has been released to the Clinch River has become incorporated in the bottom sediment. This assumption has been confirmed by the results of the inventory of radioactivity in Clinch River bottom sediment (see Retention Factors).

Too little is known about the chemical form of ^{106}Ru in the Clinch River to permit definite inferences concerning the manner in which it is incorporated in Clinch River bottom sediments. The ^{106}Ru content of annual samples of the surface layer of Clinch River bottom sediment has been shown to reflect the amount of ^{106}Ru released to the river during the preceding year.[4] Analyses of water samples from White Oak Lake indicate that less than 10 percent of the ^{106}Ru released to the Clinch River is associated with suspended sediment greater than 0.7 micron in diameter.[17] Even with the benefit of this information, it can be stated only that the mechanism by which ^{106}Ru is incorporated in the sediment may be one or more of the following: (1) sedimentation of radioactive solids suspended in White Oak Creek water, (2) precipitation of a Ru-bearing compound from Clinch River water, (3) an ion-exchange reaction between Clinch River water and bottom sediment in the river. It has been suggested[4, 23] that some of the ^{106}Ru in bottom sediments may be present in the form of nitrosyl ruthenium hydroxide-- $RuNO(OH)_3(H_2O)_2$. This compound, which is presumed to have been formed as a result of the high nitrate content of the original waste solutions, is not easily decomposed.

The general pattern of variation in gross gamma radioactivity with depth that occurs in many of the longer sediment cores taken from the Clinch River implies a similar history of sediment deposition at at each of those coring sites. Furthemore, the resemblance of the pattern, which is due primarily to the ^{137}Cs content of the sediment, to variations in the annual release of ^{137}Cs from White Oak Creek to the Clinch River, implies more or less regular, persistent deposition at those coring sites.

Regular, persistent deposition at certain coring sites in a cross section does not necessarily require regular, persistent deposition over the entire cross section. Periodic short-term net sediment losses for the cross section are possible. However, a long-term net gain for the entire period of deposition of radioactive sediment is implied for all cross sections at which cores showing the characteristic pattern were collected. This sedimentation model is confirmed by the Tennessee Valley Authority's record of sedimentation for the reaches of the Clinch River from which the cores were collected. The record shows net losses for some of the reaches downstream from CRM 13 for some of the 5-year intervals between surveys, but a net increase in sediment was computed for all reaches for the entire period of record. This net accumulation of sediment in the downstream reaches of the Clinch River is no doubt the result of readjustment of channel shape caused by the filling of Watts Bar Lake, which began at about the same time as release of radionuclides to the Clinch River was started by the Oak Ridge National Laboratory.[24]

It should be emphasized that regular and persistent sediment deposition can occur without continual sediment deposition. Because the radioactive portion of the sediment cores represents a sedimentation history of approximately 20 years, conclusions pertaining to sedimentation are at best valid for time intervals of a year or more. Sedimentation would be expected to be irregularly distributed throughout the year.

Longitudinal Distribution of Radioactive Sediment

Ninety-five percent of the radioactive bottom sediment in

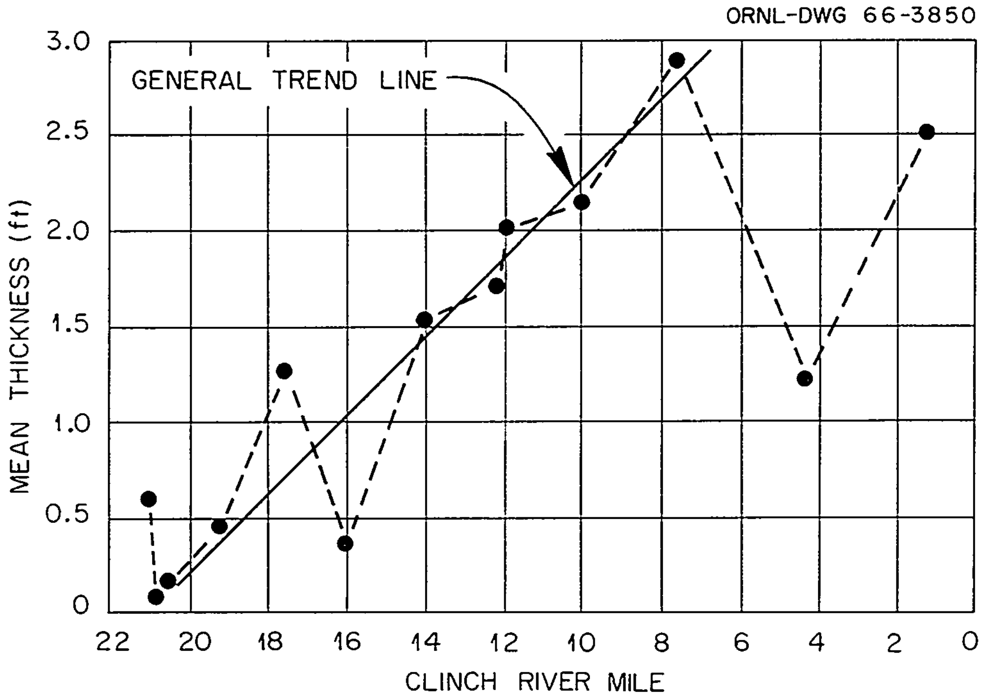

Fig. 8. Variation of Radioactive Sediment Thickness in the Downstream Direction

the Clinch River lies downstream from CRM 15 and at least 50 percent lies downstream from CRM 8.7 (Table 2).

A tendency for mean thickness of radioactive sediment to increase linearly from the head of the study reach to the mouth of the Emory River (CRM 21.0 to 4.5) appears to exist (Fig. 8). The general trend for increasing thickness may be explained by considering hydraulic conditions in the study reach. Accretions to flow in the study reach are almost negligible except inflow from the Emory River.[25] Flow area continuously increases in the downstream direction.[26] Without accretions to flow and with increasing flow area, velocity and intensity of turbulence of the stream decreases, and consequently sediment transport capacity decreases in the downstream direction.

The mean thickness of radioactive sediment is greater at the sampling section upstream from the mouth of Emory River, at CRM 7.5, than it is at the section downstream from the mouth of the river, at CRM 4.3. This difference in sediment thickness may be the result of a localized increase in turbulence downstream from the Emory River due to inflow of the river into the Clinch River and/or to the diversion of Clinch River water upstream into the Emory River.[27] An increase in turbulence would tend to keep sediment particles in suspension.

At the mouth of Poplar Creek (CRM 12.0), the mean thickness of radioactive sediment sharply increases in relation to upstream thicknesses. Probably the increased thickness, as well as the decrease in radionuclide concentrations observed in this part of the stream bed (see Fig. 10), are due to dilution of radioactive sediment by uncontaminated sediment issuing from the creek.

The section near which sediment transport capacity rapidly decreases is clearly indicated to be CRM 14.0 (Fig. 9). From CRM 21.0

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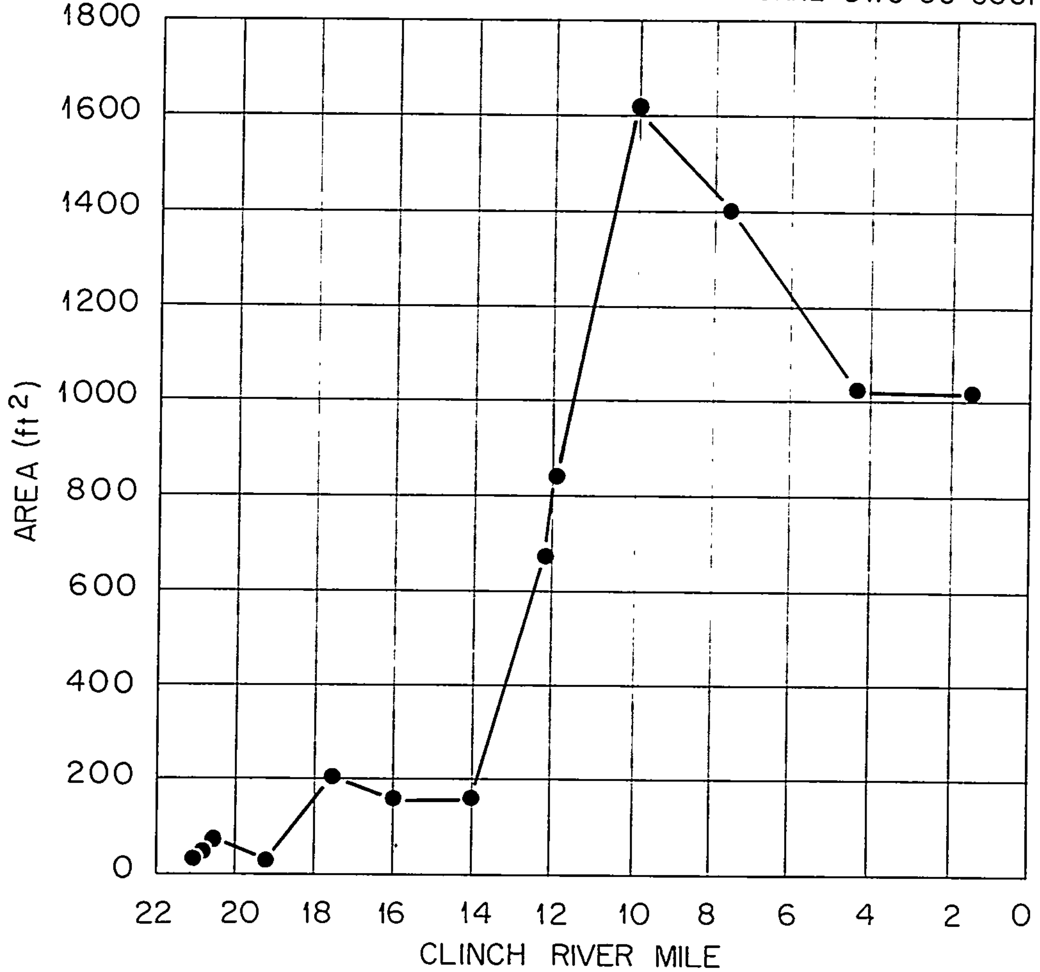

Fig. 9. Variation of Cross-Sectional Area of Radioactive Sediment in the Downstream Direction

to 14.0 deposition is limited to zones of the channel near bank(s) of the river. The sediment transport capacity is greater in the central core of flow in this reach than near the banks. Though transport capacity remains greatest in the central core of flow throughout the study reach, its magnitude relative to that near the banks drops sharply in the vicinity of CRM 14.0.

The cross-sectional area of radioactive sediment is at a maximum at CRM 10.0 (Fig. 9) because of the influence of islands and submerged ridges on the flow pattern at that cross section.

During the 19 1/2-year period of waste disposal by ORNL (December 1943 to July 1962), 64 percent of the stream bed has become covered with radioactive sediment. Areal distribution of radioactive sediments, for selected thicknesses, is listed in Table 7.

Table 7. Areal Distribution of Radioactive Sediments for Selected Thicknesses

Radioactive Sediment Thickness, in Feet	Proportion of Radioactive Surface Area Over Which Indicated Thickness is Equalled or Exceeded
1.0	0.57
1.5	.41
2.0	.31
2.6	.24
4.0	.13

Longitudinal Distribution of Radionuclides

Patterns of longitudinal variation in mean concentration are similar, in most respects, for the principal radionuclides in the sediment (Fig. 10). Highest mean concentrations of the radionuclides are in the two sections immediately downstream from the mouth of White Oak Creek (CRM 20.8 and CRM 20.5). Lowest concentrations generally are in sediments of the next downstream sampling section, at CRM 19.2. Secondary peaks in the distribution (Fig. 10) occurred at CRM 17.5 and 14.0, and for ^{106}Ru and rare earths, at CRM 4.3. Mean sectional concentrations for all but rare earths abruptly decreased between CRM 12.1 and CRM 11.9, immediately upstream and downstream from the mouth of Poplar Creek.

Distribution of several of the principal radionuclides in Clinch River bottom sediment appears to be controlled primarily by sedimentation of suspended radioactive sediment which has passed into the river through White Oak Dam.[28] Patterns of radioactive deposits in the river result from the combined interaction of lateral diffusion, characteristics of the flow pattern, and decreasing downstream intensity of turbulent forces acting to suspend the radioactive sediment particles. Therefore, similarities in concentration distribution are to be expected.

Investigation of the movement of sediment has not been a part of the Clinch River Study. As a consequence, careful documentation of the effects of diffusion, flow distribution, and turbulence on sedimentation is not available. However, the influence of these three characteristics of water movement on the longitudinal distribution of radioactive sediment may be surmised from observation and application of principles of mechanics of fluid motion.

High concentrations of radioactivity in bottom sediment may be expected near the mouth of White Oak Creek. Dispersion of waters and suspended sediments from the creek into the river is restricted because of incomplete lateral diffusion.[3, 28, 29] Consequently, suspended sediments from White Oak Creek, which are rich in sorbed radioactivity, do not immediately become diluted in the rela-

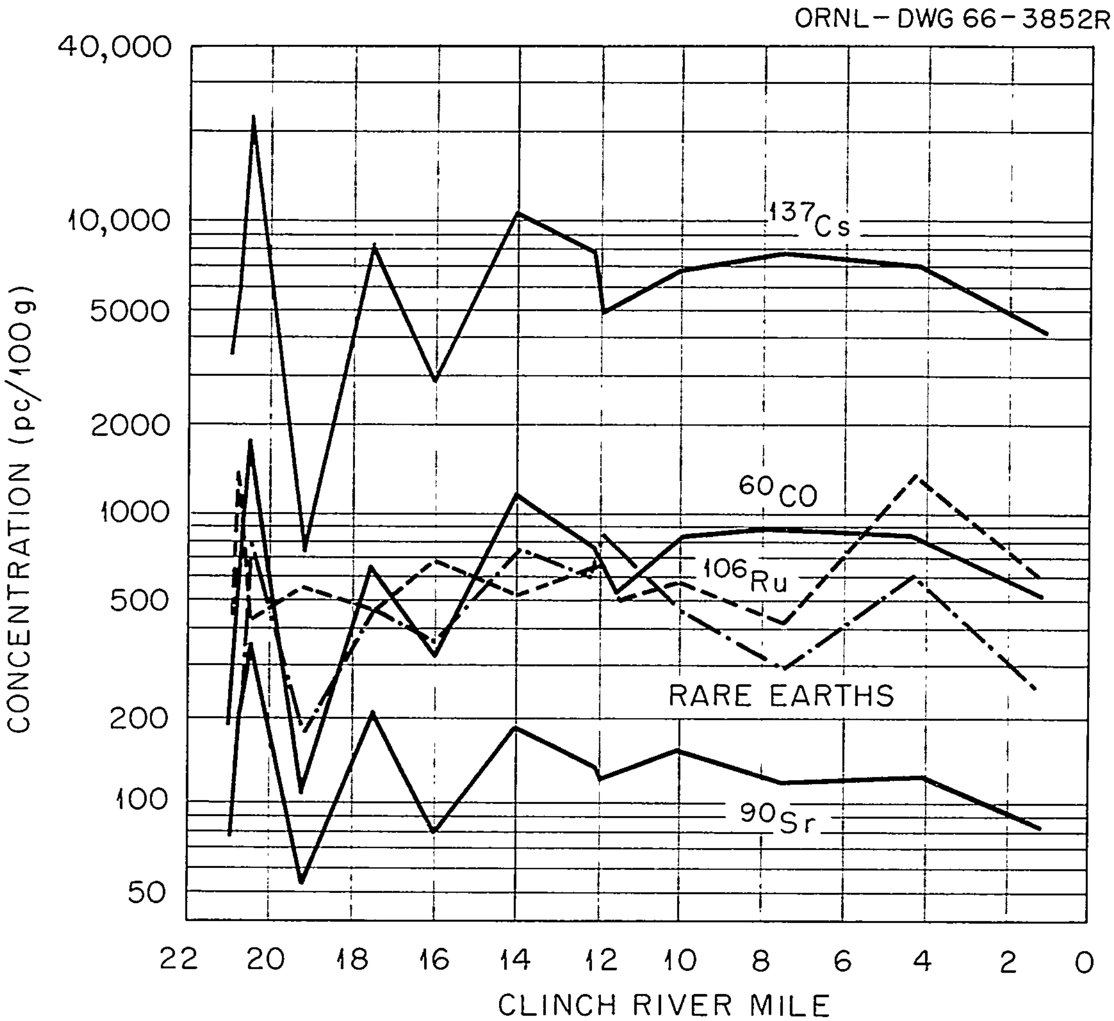

Fig. 10. Longitudinal Distribution of Radionuclides in Bottom Sediment of the Study Reach.

tively uncontaminated river.

Abrupt changes in the momentum of water entering the river from the creek tend to induce deposition. Eddy zones along the right bank of the river immediately downstream from the creek mouth are created as the two streams meet. The eddies and the restricted dispersion of suspended sediment produce immediate deposition of sediment rich in sorbed radioactivity.

As White Oak Creek water moves downstream, suspended sediment from the creek diffuses outward into the river channel, and the radioactive sediment mixes with sediment carried in the river, thus preventing formation of additional pockets of deposited sediment containing high concentrations of radioactivity.

Slightly farther downstream, turbulence is of sufficient intensity to keep the sediment in suspension through reaches such as those near CRM 19.2.

The abrupt downstream decrease in concentration of radionuclides in bottom sediment at the mouth of Poplar Creek may be the result of dilution of contaminated river sediment by uncontaminated sediment from Poplar Creek.

Relationship of Radionuclide Concentration to Thickness

Radionuclide concentrations are related to sediment thickness and location in the study reach. This relationship is shown in Figure 11. The abscissa in the figure is the ratio of the thickness at a coring vertical to mean thickness in the sampling section (shown in Fig. 8); the ordinate is the ratio of the concentration at a coring vertical to mean concentration in the sampling section (shown in Fig. 10).

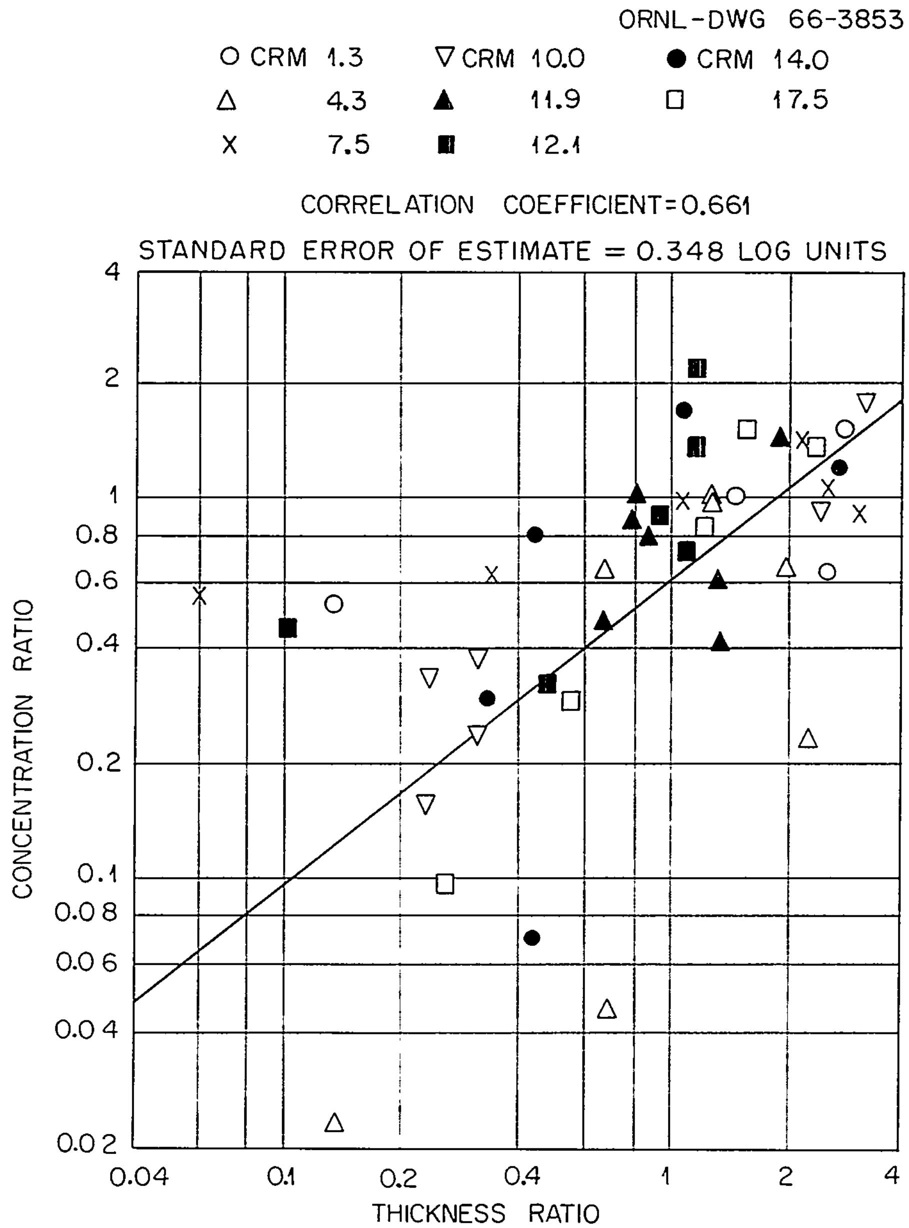

Fig. 11. Relationship of Radionuclide Concentration to Sediment Thickness

FUTURE ESTIMATION OF RADIONUCLIDE LOADING

Continued decline of radionuclide releases to the Clinch River in recent years (see Table 3) has indicated that increasing radiation hazard in the future is unlikely. Cowser and Snyder[30] find that even if releases had continued indefinitely at their highest levels (in 1956 or 1959), bottom sediment would be a minor source of radiation exposure. With continuing decline of releases, further estimation of radionuclide loading in the sediment is not needed.

Possibly waste disposal processes or quantities of radionuclides processed at ORNL will change such that increased releases to the river will result. If releases of ^{137}Cs, ^{60}Co, and/or rare earths should increase substantially above experienced levels for sustained periods, another safety analysis of radiation dosage from bottom sediment would be needed.

Safety analysis may be undertaken at two times: (1) after a period of increased loading, to determine resultant radiation dosage and (2) prior to increased loading to estimate the concentration and duration of releases which will assure safety.

For case (1), knowledge of retention factors would be an aid in estimating the magnitude of radiation dosage resulting from increased releases. The relationship of radiation dose to radioactivity of releases is

$$D = 1/2(\, 51.2\, k_r\, \bar{C}\, E\, f)$$

in which D = dose rate, in rad per day, for beta or gamma radiation from an infinitely thick source uniformly spread over the bed of the study reach,

k_r = retention factor for radionuclide, dimensionless,

$\bar{C}$ = mean radionuclide concentration for releases of known duration through White Oak Dam, in microcuries per gram (10^{-6} curies per gram),

E = effective absorbed radiation of a beta disintegration or maximum energy of gamma ray emission, in million electron volts, and

f = fraction of disintegration of a particular energy, dimensionless.

This equation for dose rate is derived from those proposed by Cowser and Snyder[30] (eqns. (14) and (17)).

D is a nominal dose rate applicable to the study reach as a whole. As such it is an indicator of relative hazard. If D approaches one-third of the critical dose rate, local dose rates may be approaching critical levels. Especially critical areas of river bed are along the right bank between CRM 20 and 21, and also in the vicinity of CRM 4.3, 14.0, and 17.5.

For case (2), safety analysis would seek an estimate of the rate of build-up of radiation to the critical dose rate. Radiation dose at the surface of the sediment increases, in general, as the radioactive sediment becomes thicker. If the radionuclide releases are constant this relationship between radiation dose and thickness is an exponential function. Cottrell[2] indicates that the dose asymptotically approaches a maximum in Clinch River bottom sediment at a thickness of about 2.6 feet; Sayre and Hubbell[31] find this asymptote is somewhat less than 2.6 feet. This thickness (2.6 feet) is called the infinite thickness; its magnitude varies slightly with variations in physicochemical properties of sediment and with energy distribution of gamma ray emissions.

Rate of build-up of radiation dose depends on the rate of accumulation of radioactive deposits; rate of accumulation, in turn, depends on location in the study reach.

Data required to estimate radiation dose would be rate of accumulation, or time to attain infinite thickness, and the interrelationship of radionuclide concentrations in releases and sediments throughout the study reach.

Information on sediment distribution is obtained from Table 7 and from Figure 8. Time required to attain an overburden of infinite thickness may be estimated by assuming the rate of accumulation at a point on the river bed is constant. The rate is assumed to be equal to the ratio of the radioactive thickness observed at the point in 1962 to the period of radionuclide releases from White Oak Creek, 19.5 years. Using this estimate of the rate of accumulation and the data in Table 7, the time required to attain infinite thickness over various proportions of the river bed may be determined.

The inter-relationship of radionuclide concentrations in releases and in sediment may be derived from the relationship of concentration and thickness (Fig. 11).

PHYSICOCHEMICAL COMPOSITION OF BOTTOM SEDIMENT

The results of physical and chemical analyses of 23 sediment samples from the two cores selected for detailed study are listed in Table 8. In general, radioactive bottom sediment in the Clinch River may be classed, according to particle size and mineralogical composition, as clayey silt[32] composed of approximately 35 percent mica and clay minerals and 65 percent quartz. The mineralogical compositions and cation exchange capacities of all 23 samples studied showed surprisingly little variation.

The mean specific bulk weight for all core composites used for the radionuclide inventory was 1.6 g/ml. The mean dry-to-wet weight ratio for all composites was 0.69.

Physical Characteristics

Particle-size distribution.--The results of particle-size analyses of the 23 samples are shown in Tables 8 and 9 and Figures 12 and 13. A diameter of 4 microns was used as the lower limit of silt-size particles in the two figures for the purpose of classification of the sediment according to particle size, which is normally based on hydraulic characteristics. A lower limit of 2 microns was used for the summarized data presented in table 8 (columns 2-7) and in constituent correlation studies, because it was believed that additional data on the finer size range would be more useful in relating particle-size distribution to the physical and chemical characteristics of the sediment. Lack of agreement between the two sets of particle-size data (columns 2-4 and columns 5-7, table 8) is the result of the use of two different methods of size analysis.

The results of the particle-size analyses by wet seiving and bottom withdrawal tube, plotted on triangular sand-silt-clay diagrams[32], are shown in Figures 12 and 13. Most of the samples fall in the clayey silt portion of the diagram.

Particle-size analyses were made also of composite samples of 45 Clinch River bottom sediment cores. Figure 14 is a plot of the results of these analyses. It is apparent that the particle-size

Table 8. Physical and Chemical Characteristics of Selected Samples of Bottom Sediment from the Clinch River[a]

Depth Interval (in.)	Particle-Size Distribution[b] (%) Sand	Silt	Clay[c]	Particle-Size Distribution[d] (%) Sand	Silt	Clay[c]	Adsorbed Water (%)	Cation Exchange Capacity (meq/100 g)	Leachable Cation Content (meq/100 g) Ca	Mg	Carbon Content (%) Mineral	Organic	Total	Free Oxide Content (%) Fe_2O_3	Al_2O_3	Minor Element Content (%) K	Rb	Cs	Sr	Radionuclide Content (pc/g)[e] ^{90}Sr	^{137}Cs	^{60}Co	^{106}Ru
											CRM 7.5, Hole No. 7-2												
10–12	40	48	12	55	34	11	34.2	12.6	21.6	8.2	0.35	2.2	2.55	1.89	0.93	1.67	0.0067	0.00044	0.0093	~0.45	37.07	3.891	6.738
14–16	28	58	14	59	28	13	34.0	12.3	18.2	5.7	0.28	2.5	2.75	1.78	0.92	1.47	0.0069	0.00031	0.0097	0.81	57.39	5.370	5.628
18–20	18	66	16	33	48	19	32.4	12.2	19.5	5.6	0.62	1.6	2.22	2.22	1.28	1.98	0.0097	0.00047	0.0105	2.0	58.28	4.682	5.476
22–24	16	63	21	23	58	19	35.1	15.1	23.7	5.6	0.15	2.4	2.50	2.32	1.41	1.96	0.0096	0.00038	0.0118	2.4	153.7	13.55	10.45
30–32	13	68	19	16	58	25	36.2	16.3	15.0	5.4	0.14	2.8	2.97	2.51	1.52	2.02	0.0107	0.00047	0.0090	3.0	305.6	21.46	−0.2331
34–36	6	64	30	21	53	26	35.1	17.9	15.0	5.8	0.21	2.8	3.05	2.48	1.53	1.91	0.0101	0.00048	0.0081	3.0	155.8	18.87	−0.04510
40–42	8	68	24	22	47	31	39.7	15.7	15.3	6.7	0.20	2.8	2.98	2.42	1.54	1.85	0.0096	0.00040	0.0079	2.4	29.14	0.6705	0.6520
44–46	12	63	25	16	58	26	39.3	18.8	14.1	6.0	0.21	2.8	3.02	2.66	1.63	1.82	0.0098	0.00047	0.0088	3.0	34.18	1.212	0.9307
50–52	17	59	24	25	52	23	46.3	23.0	18.6	7.0	0.21	4.2	4.39	2.47	1.33	1.75	0.0096	0.00046	0.0086	1.5	17.08	4.538	0.5210
52–54	18	56	26	25	51	24	36.0	18.0	16.5	5.8	0.22	2.5	2.75	2.28	1.37	1.74	0.0095	0.00038	0.0097	0.81	25.51	3.588	0.1235
56–58	9	68	23	18	56	26	36.9	18.7	12.9	7.5	0.14	2.3	2.40	2.35	1.37	1.85	0.0099	0.00039	0.0118	0.50	60.02	0.2848	0.6987
60–62	4	65	31	5	64	31	35.3	20.0	12.7	5.0	0.08	2.2	2.28	3.03	1.87	2.01	0.0113	0.00065	0.0136	~0.38	34.61	0.2122	0.6145
62–64	6	65	29	11	59	30	36.8	16.9	11.5	7.4	0.08	2.1	2.16	2.99	1.86	1.90	0.0116	0.00046	0.0153	~0.41	51.46	0.04010	0.2155
Mean value	15.0	62.4	22.6	25.3	51.3	23.4	36.7	16.7	16.5	6.3	0.22	2.6	2.77	2.42	1.43	1.84	0.0096	0.00044	0.0103	1.6	78.44	6.028	2.443
											CRM 14.0, Hole 2												
4–6	11	69	20	15	61	24	38.6	18.3	19.4	8.2	0.11	2.2	2.35	2.68	1.50	1.89	0.0100	0.00039	0.0059	1.7	127.6	11.30	16.40
8–10	8	66	26	15	60	25	46.2	25.0	17.2	10.2	0.15	2.4	2.59	2.85	1.51	2.01	0.0112	0.00039	0.0066	4.91	123.1	15.94	7.211
12–14	6	69	25	18	57	25	40.7	18.4	13.6	8.8	0.12	2.5	2.63	2.56	1.41	1.95	0.0107	0.00038	0.0061	3.2	229.4	13.39	0.8430
16–18	13	63	24	21	41	38	38.1	17.5	15.7	8.7	0.13	2.7	2.80	2.65	1.68	1.87	0.0103	0.00045	0.0063	8.60	605.1	64.63	−2.805
20–22	28	54	18	36	46	18	37.3	14.4	11.9	7.4	0.14	3.2	3.29	2.22	1.17	1.64	0.0083	0.00052	0.0059	2.7	44.16	10.20	0.6095
24–26	13	66	21	24	51	25	39.7	17.3	12.7	7.0	0.25	2.3	2.59	2.31	1.51	1.84	0.0096	0.00041	0.0060	2.2	21.04	8.868	1.289
28–30	13	67	20	31	44	25	37.2	15.0	13.2	4.9	0.22	2.3	2.49	2.22	1.40	1.82	0.0093	0.00040	0.0055	1.3	31.64	16.30	1.045
32–34	15	61	24	32	47	21	35.8	16.4	15.1	10.2	0.36	2.4	2.73	2.30	1.38	1.87	0.0096	0.00037	0.0070	1.2	33.07	4.123	0.6315
36–38	13	64	23	24	53	23	36.1	17.4	11.8	6.7	0.32	2.2	2.53	2.20	1.38	1.74	0.0092	0.00035	0.0054	1.4	82.55	0.4458	0.3147
40–42	13	63	24	23	55	22	33.5	17.9	12.8	8.6	0.33	2.1	2.40	2.23	1.28	1.77	0.0088	0.00038	0.0058	~0.47	39.88	0.2382	0.2847
Mean value	13.3	64.2	22.5	23.9	51.5	24.6	38.3	17.8	14.3	8.1	0.21	2.4	2.64	2.42	1.42	1.84	0.0097	0.00040	0.0061	2.77	143.7	14.54	2.582

[a] Results are on a dry-weight basis, except in the case of adsorbed water. Analysts, USGS – P. D. Blackmon, H. C. Starkey, H. E. Reeder, E. J. Fennelly, I. C. Frost; ORNL – T. C. Rains.

[b] Separation of sand-size particles by wet-sieving of wet samples. Separation of silt- and clay-size particles by bottom withdrawal tube.

[c] Maximum particle size, 2 μ.

[d] Separation of sand-size particles by wet sieving of previously dried sample. Separation of silt- and clay-size particles by centrifugation.

[e] Radionuclide content in picocuries per gram of sediment.

Table 9. Results of Particle-Size Analyses of Selected Samples of Bottom Sediment from the Clinch River

Depth Interval (in.)	Percent Finer than Size Indicated, in Millimeters									
	0.002	0.004	0.008	0.016	0.031	0.062	0.125	0.250	0.500	1.000
CRM 7.5, Hole No. 7-2										
10-12	12	14	18	25	37	60	93	96	98	100
14-16	14	19	23	30	43	72	88	93	95	96
18-20	16	19	26	39	55	82	91	97	100	
22-24	21	23	34	55	72	84	91	97	98	99
30-32	19	24	36	54	72	87	92	97	100	
34-36	30	31	45	60	78	94	98	99	100	
40-42	24	28	38	50	68	91	98	99	100	
44-46	25	29	43	60	79	88	95	99	100	
50-52	24	29	39	54	69	83	89	93	96	98
52-54	26	29	40	55	70	82	88	95	98	99
56-58	23	29	42	60	76	91	96	98	99	100
60-62	31	38	52	68	86	96	97	98	99	100
62-64	29	36	51	70	84	94	97	98	99	100
CRM 14.0, Hole No. 2										
4-6	20	27	39	54	74	89	95	98	100	
8-10	26	33	44	62	79	92	96	97	98	99
12-14	25	29	42	60	78	94	97	98	99	100
16-18	24	30	43	60	75	87	93	97	99	100
20-22	18	23	33	47	60	72	84	95	100	
24-26	21	28	38	51	69	87	96	98	99	100
28-30	20	25	34	48	66	87	96	99	100	
32-34	24	27	37	51	66	85	95	97	98	100
36-38	23	30	39	49	69	87	92	98	99	100
40-42	24	27	40	52	64	87	97	98	99	100

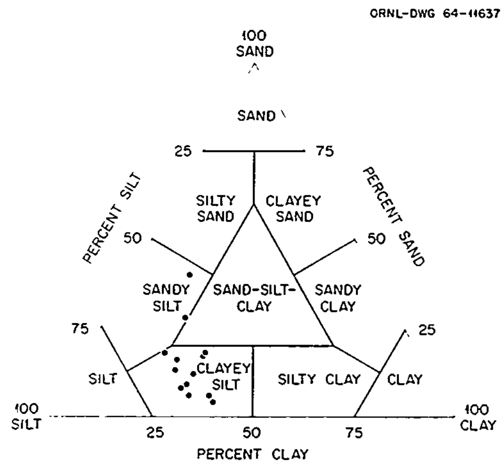

Fig. 12. Results of Particle-Size Analyses of Selected Samples from Hole 7-2, CRM 7.5. Nomenclature after Shepard[32]

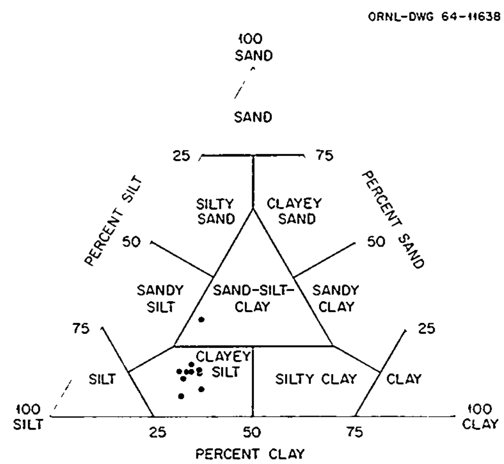

Fig. 13. Results of Particle-Size Analyses of Selected Samples from Hole 2, CRM 14.0. Nomenclature after Shepard[32]

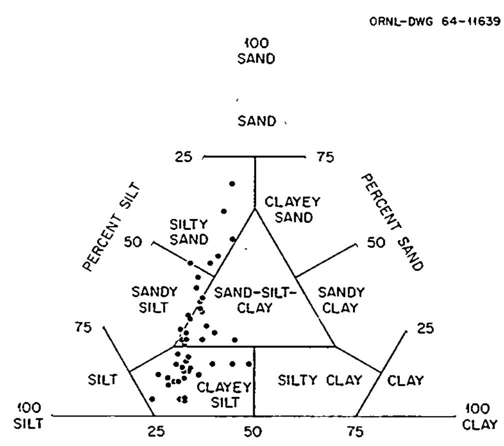

Fig. 14. Results of Particle-Size Analyses of Composite Samples of the Radioactive Portions of 45 Cores of Clinch River Bottom Sediment. Nomenclature after Shepard[32]

distributions in the composite samples, which represent the radioactive portions of cores that were taken throughout a 21-mile long reach of river, are more variable than those in the 2-inch thick incremental samples of the two cores studied in detail. Greater variations occur in the contents of sand- and silt-size particles than in the content of clay-size particles. Nevertheless, more analyses fall in the clayey silt field of the diagram than in any of the other fields. The mean contents of particles in the three size groups for all composite samples are: sand, 23 percent; silt, 54 percent; clay (< 4 microns), 23 percent. The two cores whose incremental compositions were studied in detail have average contents of sand-size particles that are still lower than the average of the composite samples (compare Figs. 12, 13, and 14).

Cation exchange capacity.--The cation exchange capacities of the unsized samples from the two cores studied in detail do not vary greatly (see Table 8). The cation exchange capacities of the clay- and silt-size fractions of the same samples do not vary greatly either (Table 10), suggesting that most of the variation in the unsized samples may be due to variations in content of clay-size material, the size fraction with the highest cation exchange capacity.

Size separations for cation-exchange capacity determinations were made by wet sieving and centrifugation after the samples had been initially dried. This procedure can result in clay-size fragments becoming attached to sand grains and may fail to disperse all aggregates of smaller grains. It can result also in concentration of organic material in the sand- and clay-size fractions. The data reported in Table 10 therefore represent the exchange capacities not of the ultimate particle-size fractions of the sediment, but of the size fractions approximately as they existed in the natural environment. The data in Table 10 are based on the same size separation as the data in columns 5-7, Table 8.

The cation exchange capacity of the clay-size sediment fraction was higher than that of the silt-size fraction in all samples, probably due to the higher content of layered aluminosilicate

Table 10. Cation Exchange Capacities of Clay-, Silt-, and Sand-Size Fractions of Selected Samples of Clinch River Bottom Sediment[1]

Depth Interval (in.)	Sand	Silt	Clay[2]	Depth Interval (in.)	Sand	Silt	Clay[2]
CRM 7.5, Hole No. 7-2				CRM 14.0, Hole 2			
10-12	n.d.[3]	11.7	42.4	4-6	n.d.	12.0	34.8
14-16	n.d.	13.8	33.5	8-10	n.d.	13.5	33.4
18-20	n.d.	9.6	37.1	12-14	n.d.	13.2	35.6
22-24	n.d.	10.2	33.2	16-18	n.d.	15.1	28.1
30-32	n.d.	10.9	33.5	20-22	n.d.	14.2	32.8
34-36	7.8	13.9	33.8	24-26	10.5	12.3	33.2
40-42	n.d.	8.9	39.2	28-30	n.d.	11.0	30.1
44-46	n.d.	14.1	33.4	32-34	n.d.	11.5	35.6
50-52	n.d.	14.9	39.1	36-38	n.d.	12.2	38.6
52-54	n.d.	12.6	30.0	40-42	n.d.	9.3	30.4
56-58	n.d.	11.6	33.4				
60-62	n.d.	10.1	34.2				
62-64	n.d.	11.1	32.2				

1 Cation exchange capacities determined by means of ammonium chloride leach and measurement of sorbed ammonium by distillation; results are in milliequivalents per 100 grams, dry weight.

2 Maximum diameter, 2 microns.

3 n.d., not determined.

minerals in the clay-size fraction of the sediment. Aluminosilicate minerals present in the other two size fractions, as well as their content of organic matter, probably account for most of the measured cation exchange capacity of these two sediment fractions.

Mineralogical Characteristics

Semi-quantitative determinations of the mineralogical compositions of the clay-, silt-, and sand-size fractions were made for all 23 sediment samples (Table 11). The size fractions studied were separated by wet seiving and centrifugation of previously dried samples, which might be expected to preserve some sand- and silt-size aggregates of smaller particles formed by drying. Careful redispersion of the sediment particles has minimized this effect, however, and the mineralogical compositions reported below are considered to be fairly representative of the three size fractions as they existed in the natural environment.

Results of the mineralogical analyses showed that the sand-size sediment fractions of all samples were composed of 70-80 percent quartz with lesser amounts of feldspar, dolomite, and mica. Kaolinite was detected in seven samples.

The silt-size fraction of each sample contained 60-70 percent quartz and lesser amounts of mica, mixed-layered mica-vermiculite, vermiculite (trioctahedral), aluminum-interlayered vermiculite (dioctahedral), kaolinite, feldspar, and dolomite (see Grim[34] for discussion of the structures of the clay minerals). Calcite was reported in three samples.

The clay-size fraction ($<$ 2 microns diameter) of each sample contained much less quartz than did the other two size fractions -- 10-20 percent. Mica and kaolinite occurred in approximately the same abundance as quartz, along with slightly lesser amounts of vermiculite (trioctahedral), aluminum-interlayered vermiculite (dioctahedral), and mixed layered mica-vermiculite. Traces of chlorite, montmorillonite, and feldspar were present in many of the samples.

Dolomite was present in the sand-size fraction of the sediment in some abundance, probably in the form of detrital grains, and was

Table 11. Mineralogical Composition of Selected Samples of Clinch River Bottom Sediment[a]

Sampling Section	Hole Number	Depth Interval (in.)	Size of Fraction	Mica	Vermiculite (Trioctahedral)	Mica-Vermiculite (Mixed-Layered)	Montmorillonite	Chlorite	Aluminum-Interlayered Vermiculite (Dioctahedral)	Kaolinite	Quartz	Feldspar	Dolomite	Calcite
CRM 7.5	7-2	10–12	Clay[b]	20	10–15	10	Trace	Trace	10–15	20	10–15	–	–	–
			Silt[c]	<10	Trace	Trace	–	–	Trace	Trace	70	<10	<10	–
			Sand[d]	<10	–[e]	–	–	–	–	–	80	<10	10	–
		14–16	Clay	20	10–15	<10	Trace	Trace	20	20	10–15	–	–	–
			Silt	10	Trace	Trace	–	Trace	–	Trace	70	Trace	Trace	Trace
			Sand	<10	–	–	–	–	–	–	80	10	<10	–
		18–20	Clay	20	10–15	<10	–	Trace	20	20	10–15	Trace	–	–
			Silt	10	Trace	Trace	–	–	Trace	<10	60–65	<10	Trace	Trace
			Sand	<10	–	–	–	–	–	–	70	10	10–15	–
		22–24	Clay	10–15	10–15	10	–	Trace	20	20	10–15	–	–	–
			Silt	<10	<10	<10	–	–	<10	<10	60–65	Trace	Trace	–
			Sand	<10	–	–	–	–	–	<10	70	<10	<10	–
		30–32	Clay	20	10–15	<10	–	Trace	20	20	10–15	Trace	–	–
			Silt	10	–	<10	–	–	Trace	Trace	60–65	10	–	–
			Sand	<10	–	–	–	–	–	–	70–75	10	10	–
		34–36	Clay	20	10–15	<10	–	Trace	10–15	20	20	Trace	–	–
			Silt	10–15	–	Trace	–	–	Trace	<10	70	<10	–	–
			Sand	Trace	–	–	–	–	–	–	70–75	10–15	10	–
		40–42	Clay	20	20	10	Trace	–	10	20	20	–	–	–
			Silt	10	–	–	–	–	–	<10	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	–	70–75	10	10	–
		44–46	Clay	20	10–15	10–15	–	Trace	10	20	10–15	–	–	–
			Silt	10	–	<10	–	–	–	Trace	70	<10	Trace	–
			Sand	<10	–	–	–	–	–	Trace	80	<10	Trace	–
		50–52	Clay	10–15	10–15	10–15	–	Trace	20	10–15	10–15	–	–	–
			Silt	10	–	–	–	10	–	Trace	70–75	Trace	Trace	–
			Sand	<10	–	–	–	–	–	–	70	10	10–15	–
		52–54	Clay	20	10–15	<10	–	Trace	20	20	10–15	–	–	–
			Silt	10	–	–	–	–	10	Trace	60–65	10	Trace	–
			Sand	<10	–	–	–	–	–	–	70–75	10	10	–
		56–58	Clay	20	10–15	10	Trace	–	10–15	20	10–15	–	–	–
			Silt	10	<10	–	–	–	<10	–	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	–	80	<10	<10	–
		60–62	Clay	20	10	10	Trace	–	20	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	<10	Trace	70	<10	Trace	Trace
			Sand	Trace	–	–	–	–	–	Trace	70–75	10–15	Trace	–
		62–64	Clay	20	10	10	–	–	20	20	20	–	–	–
			Silt	10	–	–	–	–	<10	Trace	70	10	Trace	–
			Sand	10	–	–	–	–	–	Trace	70–75	<10	<10	–

Table 11 (continued)

Sampling Section	Hole Number	Depth Interval (in.)	Size of Fraction	Mica	Vermiculite (Trioctahedral)	Mica-Vermiculite (Mixed-Layered)	Montmorillonite	Chlorite	Aluminum-Interlayered Vermiculite (Dioctahedral)	Kaolinite	Quartz	Feldspar	Dolomite	Calcite
CRM 14	2	4–6	Clay	20	10	10–15	–	Trace	10–15	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	Trace	Trace	70–75	10	–	–
			Sand	<10	–	–	–	–	–	–	80	10	Trace	–
		8–10	Clay	20	10–15	10	–	Trace	10–15	20	10–15	–	–	–
			Silt	10–15	–	Trace	–	–	Trace	<10	70	<10	–	–
			Sand	<10	–	–	–	–	–	<10	70	10	<10	–
		12–14	Clay	20	10	10–15	–	Trace	10–15	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	Trace	<10	70	<10	Trace	–
			Sand	<10	–	–	–	–	–	–	80	<10	<10	–
		16–18	Clay	20–25	10	10	Trace	Trace	10	10–15	20–25	–	–	–
			Silt	10	–	<10	–	–	<10	Trace	70	<10	Trace	–
			Sand	<10	–	–	–	–	–	–	80	10–15	–	–
		20–22	Clay	10–15	10–15	10–15	Trace	Trace	10–15	20	10–15	–	–	–
			Silt	<10	–	Trace	–	–	Trace	<10	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	–	80–85	10	<10	–
		24–26	Clay	20	10–15	10–15	–	–	10–15	20	10–15	–	–	–
			Silt	<10	–	Trace	–	–	Trace	Trace	70	10	Trace	–
			Sand	<10	–	–	–	–	–	–	70	10–15	10–15	–
		28–30	Clay	10–15	10–15	10–15	–	–	10–15	10–15	20	–	–	–
			Silt	<10	–	Trace	–	–	Trace	<10	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	Trace	70	10–15	<10	–
		32–34	Clay	10–15	10	10–15	–	–	10–15	20	20	<10	–	–
			Silt	10	–	Trace	–	–	Trace	Trace	70	<10	<10	–
			Sand	Trace	–	–	–	–	–	Trace	70–75	10	<10	–
		36–38	Clay	10–15	10–15	10–15	Trace	–	10–15	20	10–15	–	–	–
			Silt	10	Trace	Trace	–	–	Trace	Trace	70	10	Trace	–
			Sand	Trace	–	–	–	–	–	–	70–75	10	10–15	–
		40–42	Clay	10–15	10–15	10–15	–	–	20	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	Trace	–	70–75	10	<10	–
			Sand	10	–	–	–	–	–	–	70–75	10	<10	–

[a]Determined by x-ray diffraction method. Because of inherent inaccuracies of method, contents have been rounded to nearest 10%, ±5%, except where noted. Analyst, P. D. Blackmon.
[b]<2 μ in diameter.
[c]2 to 62 μ in diameter.
[d]>62 μ in diameter.
[e]Dash indicates mineral not detected.

present in the silt-size fractions of all but 4 of the 23 samples (Table 11). Calcite was detected in only a few of the samples; however, calcium carbonate can precipitate in river sediment in a poorly-crystallized form which is not susceptible to detection by X-ray methods.

The presence of precipitated calcium carbonate in the bottom sediment was of interest because, in one case where calcite precipitated from Clinch River water on a bimetallic object, the calcite contained an appreciable content of radionuclides. A radiochemical analysis of the calcite showed it to contain 123.9 pc/gram (picocuries per gram) $^{103-106}Ru$ and 10.1 pc/gram ^{90}Sr[35]. This radionuclide content is within the range of radioactivity commonly found in suspended sediment in Clinch River water downstream from the mouth of White Oak Creek. In the case cited, precipitation of the calcite may have been the result of local electrochemical action between the two metals and the river water.

In order to investigate the possible presence of poorly-crystallized calcium carbonate in Clinch River bottom sediment, the content of carbonate minerals (mineral carbon) in the raw sediment samples was determined by chemical analysis. In all samples, the content of mineral carbon was very low compared to the content of organic carbon (Table 8). The presence of dolomite as sand- and silt-size particles suggests that much of the mineral carbon content was derived from the coarser sediment fractions (Table 11). Thus, it appears that poorly-crystallized calcium carbonate is not abundant in the finer size fractions of Clinch River bottom sediment. However, its presence even in very small amounts may be significant in retention of radionuclides by the sediment.

Tamura[19] has described the strong affinity of ^{90}Sr for iron and aluminum oxides and hydroxides in basic solutions, and has suggested that this affinity is related to the surface area of the sorbent. Results of determinations of free iron oxide and free aluminum oxide in the sediment samples are listed in Table 8. Constituent correlations indicate that most of the free iron and aluminum oxides occurred in association with clay-size particles.

Organic carbon contents of the samples are listed in Table 8, also. The variation in content of organic carbon between samples is small.

Chemical Characteristics

Concentrations of one minor element and three trace elements in the sediment samples are shown in Table 8. Concentrations of potassium and rubidium were measured as possible indicators of the content of mica-type silicate minerals in the samples. The contents of cesium and strontium in the samples were of interest because in chemical sorption and ionic substitution reactions, the amount of the radioactive form of an element that becomes associated with the sediment depends on the relative abundance of the stable form of the element; for example, stable cesium will compete with radiocesium (^{137}Cs) for cation exchange sites on clay-type minerals. In the case of strontium, not only does radiostrontium (^{90}Sr) compete with stable strontium for exchange sites, but both forms compete with the much more abundant calcium. Nelson[36] measured the radiostrontium : stable strontium ratio in clam shells in the Clinch River and the Tennessee River, and reported that the ratio decreased with distance downstream from the mouth of White Oak Creek in the same manner as that predicted on the basis of flow dilution. This relationship is apparently the result of the proportional substitution of radiostrontium and stable strontium for some of the calcium ions in the calcium carbonate of the clam shell as the shell is formed.

In 22 of the 23 sediment samples, the contents of leachable calcium and magnesium total more than the cation exchange capacity of the sediment (Table 8). This observation suggests that interstitial water contained in the sediment, and minerals dissolved by the leaching solution, as well as sorbed ions displaced by ammonium ions from the leaching solution, contributed to the cation content of the leachate. The only two major leachable cations found in the sediment were calcium and magnesium. No sodium or potassium was detected. This is to be expected in the case of samples from a humid area in which the predominant rock types are limestone and dolomite, and in which the predominant cations in river water are calcium and magnesium[34].

Results of radiochemical analyses of the 23 sediment samples are shown in Table 8.

Correlations of Physicochemical Characteristics and Radionuclide Content

In general, correlations of patterns of variation in constituent concentration with depth in each of the two sediment cores studied (Tables 12 and 13) confirm the inter-relationships between cation exchange capacity, particle size, and mineralogical composition that have been described previously. The correlations also emphasize the close association of potassium and rubidium in geochemical processes. The good correlation of free iron oxide with rubidium in Hole 7-2, CRM 7.5, and with both potassium and rubidium in Hole 2, CRM 14.0, may be due to a similarity in distribution of particle sizes of free iron oxide and the minerals in which the potassium and rubidium occur. Correlation of cation exchange capacity with particle-size distribution is better when the particle-size data used are those which were obtained by chemical dispersion and analysis by bottom withdrawal tube. This method of separation should have resulted in the inclusion of essentially all clay minerals in the clay-size sediment fraction.

Mutual correlations of the contents of ^{90}Sr, ^{137}Cs, and ^{60}Co, which occur in both cores, are best in Hole 2. For Hole 7-2, a correlation of ^{137}Cs and ^{60}Co concentrations with content of particles less than 2 microns in diameter was detected after application of a correction for variations in annual releases of ^{137}Cs to the river (see Relationship of Vertical Distribution to Annual Releases). The correction was possible for only the upper five samples out of the thirteen samples analyzed in Hole 7-2. Application of a similar correction to data for all samples from Hole 2 did not materially improve the correlations. The corrected concentrations of ^{137}Cs and ^{60}Co correlate with cation exchange capacity and the contents of free

Table 12. Coefficients of Corre

	Particle-Size Distribution[a]			Particle-Size Distribution[c]			Adsorbed Water	Cation Exchange Capacity	Leachable Cation Content		Miner
	Sand	Silt	Clay[b]	Sand	Silt	Clay[b]			Ca	Mg	
Sand		−0.84	−0.85	0.91	−0.81	−0.93	−0.23	−0.57	0.71	0.33	0.5
Silt	−0.84		0.43	−0.70	0.71	0.60	0.06	0.21	−0.53	−0.36	−0.2
Clay[b]	−0.85	0.43		−0.83	0.87	0.90	0.32	0.74	−0.67	−0.20	−0.6
Sand	0.91	−0.70	−0.83		−0.97	−0.91	−0.29	−0.67	0.64	0.25	0.5
Silt	−0.81	0.62	0.75	−0.97		0.77	0.23	0.64	−0.48	−0.28	−0.5
Clay[b]	−0.93	0.71	0.87	−0.91	0.77		0.36	0.63	−0.79	−0.16	−0.5
Adsorbed water	−0.23	0.06	0.32	−0.29	0.23	0.36		0.74	−0.16	−0.29	−0.
Cation exchange capacity	−0.57	0.21	0.74	−0.67	0.64	0.63	0.74		−0.47	−0.001	−0.
Calcium	0.71	−0.53	−0.67	0.64	−0.48	−0.79	−0.16	−0.47		0.01	0.
Magnesium	0.33	−0.36	−0.20	0.25	−0.28	−0.16	0.29	−0.001	0.01		−0.0
Mineral carbon	0.52	−0.25	−0.63	0.58	−0.53	−0.58	−0.35	−0.59	0.51	−0.01	
Organic carbon	−0.07	−0.09	0.20	−0.09	0.06	0.14	0.88	0.66	0.02	−0.09	−0.
Total carbon	0.05	−0.16	0.07	0.04	−0.07	0.01	0.83	0.55	0.13	0.08	−0.
Fe_2O_3	−0.83	0.56	0.84	−0.92	0.87	0.87	0.29	0.64	−0.69	−0.18	−0.
Al_2O_3	−0.89	0.64	0.86	−0.94	0.88	0.91	0.20	0.55	−0.69	−0.26	−0.
Potassium	−0.67	0.69	0.45	−0.78	0.83	0.59	−0.09	0.23	−0.25	−0.35	−0.
Rubidium	−0.90	0.74	0.79	−0.96	0.92	0.87	0.19	0.56	−0.64	−0.29	−0.
Cesium	−0.44	0.23	0.50	−0.58	0.59	0.49	0.04	0.43	−0.39	−0.26	−0.
Strontium	−0.31	0.21	0.31	−0.37	0.42	0.26	−0.27	0.05	−0.30	0.04	−0.
^{90}Sr	−0.15	0.28	−0.01	−0.17	0.27	−0.04	−0.27	−0.13	0.11	−0.53	0.
^{137}Cs	−0.15	0.36	−0.10	−0.18	0.25	0.003	−0.25	−0.14	0.05	−0.38	−0.
^{60}Co	−0.01	0.13	−0.10	−0.002	0.08	−0.16	−0.20	−0.12	0.26	−0.41	−0.
^{106}Ru	0.61	−0.37	−0.66	0.59	−0.42	−0.76	−0.45	−0.66	0.86	−0.02	0.
^{137}Cs corrected[d]	−0.62	0.55	0.68	−0.77	0.71	0.81	0.89	0.97	−0.49	−0.40	−0.
^{60}Co corrected[d]	−0.60	0.49	0.74	−0.76	0.73	0.75	0.93	0.99	−0.34	−0.38	−0.

[a]Separation of sand-size particles by wet sieving of wet sample. Determination of content of silt- and clay-siz

[b]Maximum particle diameter, 2 μ.

[c]Separation of sand-size particles by wet sieving of previously dried sample. Separation of silt- and clay-size

[d]Five values only.

on for Constituent Pairs, Hole 7-2, CRM 7.5

rbon Content		Free Oxide Content		Minor Element Content				Radionuclide Content					
Organic	Total	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	^{90}Sr	^{137}Cs	^{60}Co	^{106}Ru	^{137}Cs Corrected	^{60}Co Corrected
−0.07	0.05	−0.83	−0.89	−0.67	−0.90	−0.44	−0.31	−0.15	−0.15	−0.01	0.61	−0.62	−0.60
−0.09	−0.16	0.56	0.64	0.69	0.74	0.23	0.21	0.28	0.36	0.13	−0.37	0.55	0.49
0.20	0.07	0.84	0.86	0.45	0.79	0.50	0.31	−0.01	−0.10	−0.10	−0.66	0.68	0.74
−0.09	0.04	−0.92	−0.94	−0.78	−0.96	−0.58	−0.37	−0.17	−0.18	−0.002	0.59	−0.77	−0.76
0.06	−0.07	0.87	0.88	0.83	0.92	0.59	0.42	0.27	0.25	0.08	−0.42	0.71	0.73
0.14	0.01	0.87	0.91	0.59	0.87	0.49	0.26	−0.04	0.003	−0.16	−0.76	0.81	0.75
0.88	0.83	0.29	0.20	−0.09	0.19	0.04	−0.27	−0.27	−0.25	−0.20	−0.45	0.89	0.93
0.66	0.55	0.64	0.55	0.23	0.56	0.43	0.05	−0.13	−0.14	−0.12	−0.66	0.97	0.99
0.02	0.13	−0.69	−0.69	−0.25	−0.64	−0.39	−0.30	0.11	0.05	0.26	0.86	−0.49	−0.34
0.09	0.08	−0.18	−0.26	−0.35	−0.29	−0.26	0.04	−0.53	−0.38	−0.41	−0.02	−0.40	−0.38
−0.32	−0.09	−0.58	−0.58	−0.17	−0.47	−0.17	−0.34	0.12	−0.19	−0.04	0.42	−0.71	−0.81
	0.97	0.06	−0.01	−0.22	−0.006	−0.06	−0.52	−0.05	0.01	0.18	−0.35	0.74	0.77
0.97		−0.07	−0.14	−0.26	−0.11	−0.08	−0.65	−0.004	−0.02	0.19	−0.29	0.71	0.69
0.06	−0.07		0.97	0.68	0.92	0.72	0.51	0.04	0.03	−0.14	−0.58	0.79	0.76
−0.01	−0.14	0.97		0.72	0.94	0.64	0.46	0.12	0.10	−0.08	−0.57	0.77	0.75
−0.22	−0.26	0.68	0.72		0.82	0.62	0.32	0.40	0.46	0.30	−0.19	0.56	0.53
−0.006	−0.11	0.92	0.94	0.82		0.59	0.47	0.17	0.24	0.06	−0.56	0.69	0.65
−0.06	−0.08	0.72	0.64	0.62	0.59		0.30	0.06	0.02	−0.04	−0.36	0.30	0.20
−0.52	−0.65	0.51	0.46	0.32	0.47	0.30		−0.30	−0.13	−0.35	0.07	−0.19	−0.07
−0.05	−0.004	0.04	0.12	0.40	0.17	0.06	−0.30		0.69	0.72	0.11	0.78	0.76
0.01	−0.02	0.03	0.10	0.46	0.24	0.02	−0.13	0.69		0.90	0.03	0.99	0.96
0.18	0.19	−0.14	−0.08	0.30	0.06	−0.04	−0.35	0.72	0.90		0.12	0.99	0.98
−0.35	−0.29	−0.58	−0.57	−0.19	−0.56	−0.36	0.07	0.11	0.03	0.12		−0.54	−0.39
0.74	0.71	0.79	0.77	0.56	0.69	0.30	−0.19	0.78	0.99	0.99	−0.54		0.98
0.77	0.69	0.76	0.75	0.53	0.65	0.20	−0.07	0.76	0.96	0.98	−0.39	0.98	

articles by bottom withdrawal tube.

rticles by centrifugation.

Table 13. Coefficients

	Particle-Size Distribution[a]			Particle-Size Distribution[c]			Adsorbed Water	Cation Exchange Capacity	Leachable Cation Content	
	Sand	Silt	Clay[b]	Sand	Silt	Clay[b]			Ca	Mg
Sand		−0.91	−0.69	0.80	−0.52	−0.40	−0.43	−0.63	−0.42	−0.26
Silt	−0.91		0.33	−0.74	0.51	0.32	0.37	0.43	0.43	−0.06
Clay[b]	−0.69	0.33		−0.53	0.28	0.34	0.32	0.68	0.20	0.68
Sand	0.80	−0.74	−0.53		−0.72	−0.42	−0.55	−0.77	−0.63	−0.38
Silt	−0.52	0.51	0.28	−0.72		−0.32	0.41	0.67	0.43	0.34
Clay[b]	−0.40	0.32	0.34	−0.42	−0.32		0.20	0.19	0.31	0.07
Adsorbed water	−0.43	0.37	0.32	−0.55	0.41	0.20		0.77	0.45	0.32
Cation exchange capacity	−0.63	0.43	0.68	−0.77	0.67	0.19	0.77		0.54	0.59
Calcium	−0.42	0.43	0.20	−0.63	0.43	0.31	0.45	0.54		0.50
Magnesium	−0.26	−0.06	0.68	−0.38	0.34	0.07	0.32	0.59	0.50	
Mineral carbon	0.07	−0.20	0.17	0.41	−0.14	−0.36	−0.60	−0.23	−0.45	−0.04
Organic carbon	0.69	−0.71	−0.34	0.46	−0.49	−0.005	0.12	−0.30	−0.19	0.05
Total carbon	0.78	−0.83	−0.32	0.60	−0.54	−0.13	0.009	−0.35	−0.34	0.07
Fe_2O_3	−0.55	0.46	0.45	−0.79	0.45	0.50	0.77	0.77	0.82	0.58
Al_2O_3	−0.60	0.56	0.38	−0.62	−0.02	0.87	0.44	0.42	0.57	0.19
Potassium	−0.84	0.22	0.66	−0.73	0.44	0.41	0.71	0.74	0.67	0.54
Rubidium	−0.79	0.65	0.67	−0.77	0.39	0.52	0.80	0.78	0.64	0.52
Cesium	0.76	−0.67	−0.56	0.44	−0.49	0.05	0.002	−0.36	−0.16	−0.14
Strontium	−0.12	−0.13	0.50	−0.08	−0.01	0.12	0.35	0.39	0.46	0.85
^{90}Sr	−0.12	−0.03	0.33	−0.33	−0.29	0.82	0.47	0.33	0.31	0.31
^{137}Cs	−0.28	0.12	0.43	−0.45	−0.22	0.90	0.30	0.29	0.38	0.33
^{60}Co	−0.08	0.02	0.15	−0.20	−0.50	0.91	0.20	0.05	0.30	0.10
^{106}Ru	−0.27	0.44	−0.16	−0.58	0.66	−0.05	0.37	0.41	0.84	0.18
^{137}Cs corrected	−0.23	0.09	0.36	−0.41	−0.27	0.91	0.27	0.24	0.35	0.27
^{60}Co corrected	−0.08	0.02	0.14	−0.19	−0.50	0.91	0.20	0.05	0.30	0.09

[a] Separation of sand-size particles by wet sieving of wet samples. Determination of content of silt- and

[b] Maximum particle diameter, 2 μ.

[c] Separation of sand-size particles by wet sieving of previously dried sample. Separation of silt- and cla

Correlation for Constituent Pairs, Hole 2, CRM 14.0

Carbon Content			Free Oxide Content		Minor Element Content				Radionuclide Content					
eral	Organic	Total	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	^{90}Sr	^{137}Cs	^{60}Co	^{106}Ru	^{137}Cs Corrected	^{60}Co Corrected
.07	0.69	0.78	−0.55	−0.60	−0.84	−0.79	0.76	−0.12	−0.12	−0.28	−0.08	−0.27	−0.23	−0.08
.20	−0.71	−0.83	0.46	0.56	0.22	0.65	−0.67	−0.13	−0.03	0.12	0.02	0.44	0.09	0.02
.17	−0.34	−0.32	0.45	0.38	0.66	0.67	−0.56	0.50	0.33	0.43	0.15	−0.16	0.36	0.14
.41	0.46	0.60	−0.79	−0.62	−0.73	−0.77	0.44	−0.08	−0.33	−0.45	−0.20	−0.58	−0.41	−0.19
.14	−0.49	−0.54	0.45	−0.02	0.44	0.39	−0.49	−0.01	−0.29	−0.22	−0.50	0.66	−0.27	−0.50
.36	−0.005	−0.13	0.50	0.87	0.41	0.52	0.05	0.12	0.82	0.90	0.91	−0.05	0.91	0.91
.60	0.12	0.009	0.77	0.44	0.71	0.80	0.002	0.35	0.47	0.30	0.20	0.37	0.27	0.20
.23	−0.30	−0.35	0.77	0.42	0.74	0.78	−0.36	0.39	0.33	0.29	0.05	0.41	0.24	0.05
.45	−0.19	−0.34	0.82	0.57	0.67	0.64	−0.16	0.46	0.31	0.38	0.30	0.84	0.35	−0.30
.04	0.05	0.07	0.58	0.19	0.54	0.52	−0.14	0.85	0.31	0.33	0.10	0.18	0.27	0.09
	−0.41	−0.24	−0.68	−0.33	−0.31	−0.46	−0.50	−0.004	−0.59	−0.53	−0.52	−0.45	−0.53	−0.52
.48		0.96	0.003	−0.24	−0.35	−0.19	0.89	0.18	0.45	0.27	0.37	−0.26	0.29	0.37
.24	0.96		−0.17	−0.35	−0.46	−0.31	0.83	0.22	0.33	0.12	0.23	−0.40	0.15	0.23
.68	0.003	−0.17		0.67	0.81	0.88	−0.05	0.46	0.64	0.65	0.48	0.60	0.61	0.48
.33	−0.24	−0.35	0.67		0.67	0.73	−0.19	0.28	0.68	0.72	0.70	0.27	0.72	0.70
.31	−0.35	−0.46	0.81	0.67		0.95	−0.46	0.55	0.36	0.40	0.25	0.40	0.34	0.24
.46	−0.19	−0.31	0.88	0.73	0.95		−0.34	0.49	0.55	0.57	0.39	0.36	0.52	0.39
.50	0.89	0.83	−0.05	−0.19	−0.46	−0.34		−0.009	0.40	0.20	0.40	−0.14	0.25	0.41
.004	0.18	0.22	0.46	0.28	0.55	0.49	−0.009		0.35	0.27	0.20	0.05	0.22	0.20
.59	0.45	0.33	0.64	0.68	0.36	0.55	0.40	0.35		0.94	0.92	−0.05	0.94	0.92
.53	0.27	0.12	0.65	0.72	0.40	0.57	0.20	0.27	0.94		0.92	−0.009	0.99	0.92
.52	0.37	0.23	0.48	0.70	0.25	0.39	0.40	0.20	0.92	0.92		−0.08	0.94	$\sim$1.00
.45	−0.26	−0.40	0.60	0.27	0.40	0.36	−0.14	0.05	−0.05	−0.009	−0.08		−0.02	−0.08
.53	0.29	0.15	0.61	0.72	0.34	0.52	0.25	0.22	0.94	0.99	0.94	−0.02		0.94
.52	0.37	0.23	0.48	0.70	0.24	0.39	0.41	0.20	0.92	0.92	$\sim$1.00	−0.08	0.94	

y-size particles by bottom withdrawal tube.

ize particles by centrifugation.

iron oxide, free aluminum oxide, adsorbed water, and organic matter in Hole 7-2, and with free iron oxide content and free aluminum oxide content in Hole 2. The content of ^{106}Ru correlates with leachable calcium in both cores.

In the 45 composite samples of bottom sediment cores that were analyzed for both particle-size distribution and radionuclide content, the content of sand-size particles showed a strong negative correlation with the content of silt-size particles, emphasizing the previously-noted small variation in content of clay-size particles in the composite samples. No correlation of particle-size distribution with the content of individual radionuclides in the samples was noted. A strong correlation of ^{137}Cs concentration with ^{60}Co concentration observed in the composite samples is consistent with the correlation of those two radionuclides observed in other samples of Clinch River bottom sediment.

The quantity of a radionuclide sorbed on river sediment might be expected to increase with decreasing particle size of the sediment for two reasons: (1) the relative content of layered aluminosilicate minerals, the mineral group with the highest sorption capacities for many cations[34], usually increases as the mean particle size of the sediment decreases; (2) in some materials, including several of the layered aluminosilicate minerals and organic matter, sorption capacity varies directly with surface area, which increases as particle size decreases[34, 37].

The study of the relationship between particle-size distribution and radionuclide content of Clinch River bottom sediment is complicated by the effect of variations in the amounts of each of the radionuclides released annually. Both cores studied in detail exhibited patterns of variation in gross gamma radioactivity with depth which were similar to the pattern of annual releases of ^{137}Cs to the Clinch River from ORNL. Because ^{137}Cs and ^{60}Co account for most of the radioactivity in the sediment, the effect of annual releases must be corrected for in order to investigate the effect of particle-size distribution on the content of the two radionuclides in Clinch River bottom sediment. Attempts at such a correction were made in two ways: (1) in the incremental samples of the two sediment cores, by decreasing the measured radiochemical concentrations of the individual slices by an amount proportional to the amount of ^{137}Cs released during the years which the sample is believed to represent; (2) in the case of the composite samples of the sediment cores, by using cores from portions of the river in which there has been a great preponderance of sediment deposition over sediment erosion, thus obtaining an integrated sample representing sediment deposited during the entire period since 1943.

Another complicating factor in a study of the relationship of sediment composition to radionuclide content is the effect of dilution of radioactive sediment by non-radioactive sediment. If incorporation of ^{137}Cs and ^{60}Co in Clinch River bottom sediment is by deposition of suspended radioactive solids, as has been suggested in this report, the concentration of the two radionuclides at a given sample site, and at a given depth, will be greatly affected by the amount of non-radioactive sediment deposited with the radioactive sediment. Because the proportions of the two kinds of sediment can be expected to vary considerably from place to place, and from time to time at the same place, only a rather imperfect correlation of radionuclide content and sediment composition can be expected at best.

The lack of consistent correlations with particle-size of the corrected ^{137}Cs and ^{60}Co content of the incremental samples from the two cores is probably the result of the complicating factors just described. Corrections for the effects of annual radionuclide releases which were applied to the analyses for the two radionuclides may not have been adequate. Effects of dilution of radioactive sediment by non-radioactive sediment may have been considerable. Particle-size ranges (sand, silt, clay) used in the comparisons may not have been detailed enough to demonstrate particle size-radionuclide content relationships. The latter two statements might be used also to explain the poor correlations between particle-size and radionuclide content exhibited by the 45 composite samples analyzed.

Concentrations of radionuclides which have been incorporated in bottom sediment as a result of simple cation exchange reactions might be expected to correlate with the total cation exchange capacity of the sediment. Desorption experiments performed on sediment collected near the mouth of White Oak Creek have indicated that of the four most important radionuclides in Clinch River bottom sediment (^{90}Sr, ^{137}Cs, ^{60}Co, ^{106}Ru), only ^{90}Sr was held primarily by simple ion exchange[4]. Evidence that some of the ^{90}Sr was associated with calcium carbonate in the sediment was obtained also.

The content of ^{90}Sr in the samples from the two sediment cores does not show a good correlation with either the total cation exchange capacity or the mineral carbon (calcium carbonate) content of the samples. This lack of good correlation may be due to the effect of annual releases of ^{90}Sr. Measured concentrations of ^{90}Sr could not be corrected for variations in annual releases as were concentrations of ^{137}Cs and ^{60}Co.

In the two samples for which the cation exchange capacity of the sand-size sediment fraction was determined, the cation exchange capacity of the silt-size sediment fraction exceeds that of the sand-size fraction. In all samples the cation exchange capacity of the clay-size fraction exceeds that of the silt-size fraction by a ratio of nearly 3:1 (see Table 10). These relationships between the various size fractions of the sediment can be largely explained by the relative content of layered aluminosilicate minerals in each size fraction. The ratio of the content of these aluminosilicate minerals in the clay-size fraction to that in the silt-size fraction is also almost 3:1. The higher specific surface area of the clay-size fraction no doubt has modified the effect of mineralogy on the cation exchange capacity of the sediment.

The role which calcium carbonate has played in the incorporation of radionuclides in Clinch River bottom sediment requires further clarification. Evidence for the precipitation of calcium carbonate in the Clinch River has been obtained, yet the carbonate content of the sediment appears to be minor, and much of that which is present appears to be in the form of detrital carbonate grains associated with the larger particles of sediment. It has been observed[4] that the content of calcium carbonate is higher in bottom sediment from White Oak Lake than in bottom sediment from near the mouth of White Oak Creek, 0.6 mile downstream, and that the fraction of the total content of ^{90}Sr desorbed from White Oak Lake sediment through ion exchange mechanisms is less than in the case of sediment from near the mouth of the creek. The tendency of strontium to substitute for calcium in the lattices of calcium carbonate minerals[36]; the observed inverse relationships between the calcium carbonate content of White Oak Creek bottom sediment and the amount of ^{90}Sr held through ion exchange; the incorporation of both ^{90}Sr and ^{106}Ru in calcium carbonate that precipitated in the Clinch River, and the correlation of ^{106}Ru with leachable calcium in Clinch River bottom sediment; all illustrate the need for further study of the role which calcium carbonate plays in incorporation of radionuclides in Clinch River bottom sediment.

The significance of the content of free oxides in the samples of bottom sediment requires further investigation. The correlation of free iron oxide with the clay-size sediment fraction suggests that iron oxide coatings on sand grains are not a major source of the iron. Iron and aluminum oxides should be investigated further because of their high specific sorption capacities for strontium. Sorption of cobalt by iron and manganese oxides also should be investigated.

The content of organic matter (organic carbon) appears to add to the cation exchange capacity of the sediment in Hole 7-2 (see Table 12). The reported high cation exchange capacities of a number of organic materials[38] support this observation. Both vegetal material, consisting of individual leaves and leaf mats, twigs, and other woody material, and metabolic products and remains of aquatic animals, are present in the river. Coal is an obvious constituent of the coarser sediment fractions in many places throughout the study reach.

The use of minor and trace elements to indicate the relative abundance of certain layered aluminosilicate minerals in samples of Clinch River bottom sediment appears to be complicated by the presence of other minerals, such as feldspars, which contain the same constituents. The content of adsorbed water in the sediment seems to be a better indication of its cation exchange capacity, and possibly therefore its content of layered aluminosilicate minerals.

The mutual correlations between the contents of ^{90}Sr, ^{137}Cs, and ^{60}Co in the two cores (quite strong correlation in Hole 2) suggest that the ^{90}Sr content of Clinch River bottom sediment may be related in some way to the process of sedimentation of suspended matter in the river.

CONCLUSIONS

The inventory of radionuclides in Clinch River bottom sediment provides strong insight to the fate of radioactive waste released to the Clinch River. The inventory is a direct and principal measure of the residual between 20-year input and output loads of the radionuclides in the study reach of the river.

In the study reach at least 20 percent of the ^{137}Cs and rare earths released to the river are retained in the bottom sediment. Retention of ^{60}Co releases is 9 percent. Less than 1 percent each of ^{106}Ru and ^{90}Sr releases are retained in the bottom sediment. Most of the radioactive sediment, 95 percent, is in that portion of the channel bed between CRM 0 and CRM 15. Because of downstream decrease in the turbulence of flow, the thickness, cross-sectional area, and volume per unit length of radioactive sediment are generally greater in the downstream parts of the reach than in the upstream parts. Of these three geometric variables, only thickness varies in a regular manner, showing a linear increase in the downstream direction.

The total inventory of principal radionuclides in bottom sediment (^{137}Cs, ^{60}Co, and rare earths) in the Tennessee River basin is greater than that measured for the main channel of the Clinch River. Radionuclides are associated with bottom sediment of sloughs and mouths of streams tributary to the river. Downstream from the mouth of the Clinch River in Watts Bar Lake a large volume of sediment deposits exist and significant concentrations of the principal radionuclides are found in the upper strata of the bottom sediment (see ref. 29).

Variations in gross gamma radioactivity with depth in Clinch River bottom sediment largely reflect variations in the content of ^{137}Cs in the sediment. A similar pattern of variations in the content of ^{137}Cs in the sediment at several coring sites in the reach of river downstream from CRM 18 indicates more or less regular, persistent deposition of sediment at those particular sites, and thus longterm net accumulation of sediment at those sampling sections.

Incorporation of ^{137}Cs in Clinch River bottom sediment by sedimentation of cesium-bearing aluminosilicate minerals entering the

Clinch River from the Oak Ridge National Laboratory is indicated by: (a) the similarity of the patterns of variations with depth in ^{137}Cs content of bottom sediment cores to the pattern of annual releases of ^{137}Cs from the Laboratory to the Clinch River, (b) the knowledge that at least 70 percent of the ^{137}Cs released from the Laboratory to the Clinch River was associated with suspended solids, and (c) the known preferential sorption of cesium by certain layered aluminosilicate minerals.

The similarity of the distribution pattern of ^{60}Co to that of ^{137}Cs in Clinch River bottom sediment suggests that ^{60}Co may be incorporated in the sediment by deposition of suspended solids entering the Clinch River from White Oak Creek. The two radionuclides are not necessarily associated with the same solids, however. The fact that the distribution pattern of ^{60}Co does not reflect the pattern of annual releases of ^{60}Co from White Oak Creek implies that most of the ^{60}Co released to the Clinch River is not associated with suspended sediment large enough to be deposited in the Clinch River, and that a rather small fraction of the total ^{60}Co released to the river becomes incorporated in Clinch River bottom sediment.

The relatively small contribution of ^{106}Ru to the gross gamma radioactivity of the upper portion of Clinch River bottom sediment supports the results of the radionuclide inventory, which has shown that only a small fraction of the total amount of ^{106}Ru released to the river is incorporated in the bottom sediment. The manner of incorporation of ^{106}Ru in the sediment is not known.

Cation exchange properties of Clinch River bottom sediment are largely controlled by its content of mica and clay minerals. These minerals are found primarily in the finest sediment fraction, but occur also in the coarser fractions in the form of mineral aggregates and shale particles. Calcium and magnesium are the major leachable cations in the sediment; potassium and sodium are not present in measureable quantities.

The effects of chemical and physical properties of Clinch River bottom sediment on its radionuclide content are obscured as a result of variations in annual releases of the radionuclides, and as a result of dilution of radioactive sediment by non-radioactive sediment in the river. Corrections for these two factors should be applied if an assessment of the effect of sediment composition on radionuclide content is sought.

Two methods of aiding future safety analyses are suggested: (1) estimation of dose rates for individual radionuclides incorporated in bottom sediment by consideration of mean concentration of the radionuclides in releases and of the retention factor, and (2) consideration of (a) rate of sediment thickness build-up in various areas of the study reach, (b) relation of concentration to sediment thickness, and (c) inter-relationship of concentrations in releases and in sediments.

Future use of the river for radioactive liquid waste disposal is predicated on the safety of such practices. It has been concluded that past and present disposal practices have been safe[31], and that continued use of the river for such disposal is permissible. A projected decline in release of radionuclides to the river suggests that retention of radionuclides in bottom sediment will not seriously limit the river's usefulness in the immediate future, either. However, the possibility of an increase in radionuclide releases and, consequently, the possibility of increased radiation hazard, must be recognized. If it is assumed that retention of some radionuclides in bottom sediment will reach 10 to 20 percent, or more, the incorporation of radionuclides from a substantial increase in releases might be a factor leading to limited use of the stream for disposal of radioactive waste. Limitations resulting from such an increase in the radionuclide content of bottom sediment can be determined only through a safety analysis in which all avenues of radiation exposure are considered.

Water-sampling stations have value in obtaining continuous and current records of radionuclide concentrations at a site. However, measurement of small but significant losses or gains in radionuclide loads occurring between stations of a network is difficult and costly. Inventory of accumulated radioactivity in sediment is inherently more accurate than determining a small residual between large radionuclide loads measured at two water-sampling stations. Cost of such an inventory would be comparable to the annual cost of operating a two-station water-sampling network in the Clinch River study reach. The techniques of radioactive sediment inventory developed in this investigation can be used for surveillance of long-term effects of the release of radioactive material. The method can be used in lieu of water-sampling networks.

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SAFETY ANALYSIS OF RADIONUCLIDE RELEASE TO THE CLINCH RIVER

K. E. Cowser
W. S. Snyder

OAK RIDGE NATIONAL LABORATORY
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HEALTH PHYSICS DIVISION

SAFETY ANALYSIS OF RADIONUCLIDE RELEASE TO THE CLINCH RIVER

Supplement No. 3 to Status Report No. 5 on Clinch River Study

By

K. E. Cowser and W. S. Snyder

Progress Report No. 3 (Final) of Subcommittee on Safety Evaluation. Presented at Meeting of Clinch River Study Steering Committee, December 15-16, 1964.

MAY 1966

OAK RIDGE NATIONAL LABORATORY
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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

Acknowledgment

The Subcommittee on Safety Evaluation was established by the Clinch River Study Steering Committee October 27, 1961, to study available information and additional data that might be obtained and to evaluate the potential radiation exposure that might result from discharges of radioactive waste water from Oak Ridge installations to the Clinch and Tennessee Rivers. Previously, the Steering Committee had established subcommittees on Water Sampling and Analysis, Bottom Sediment Sampling and Analysis, and Aquatic Biology.

Membership of the Subcommittee on Safety Evaluation has included C. P. McCammon (TDPH), Chairman, O. W. Kochtitzky (TVA), C. P. Straub (USPHS), R. L. Hervin (AEC-ORO), and W. S. Snyder (ORNL). Their technical guidance and professional judgment were invaluable during the formative stage and during the conduct of the safety study. The senior author was assigned staff responsibility to this subcommittee. Descriptive and analytical data needed to define exposure factors were furnished primarily by the other subcommittees of the Clinch River Study Steering Committee and by TVA, USGS, USPHS, and the Applied Health Physics Section, Health Physics Division, ORNL.

Acknowledgment is due two staff members of the Health Physics Division. M. J. Cook was responsible for preparation of the dose correction factors shown in Figures 3, 4, and 5, and D. G. Jacobs furnished information and technical guidance in the calculation of stable and radioactive elements retained by soils. Constructive comments were made by R. F. Reitemeier, Division of Biology and Medicine, AEC, Washington, and R. G. Menzel and J. E. Ambler, Agricultural Research Service, USDA, Beltsville, on fission product transfer to crops irrigated with contaminated water.

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ABSTRACT

One of the principal objectives of the Clinch River Study has been the evaluation of radiation dose equivalents to populations downstream from operations at the Oak Ridge National Laboratory. Evaluation was based on long-term monitoring data (1944 to 1963) and the identification of critical exposure pathways and population groups.

Knowledge of water utilization downstream indicates that the important avenues of exposure from discharge of low-level contaminated waste water to the Clinch River include: 1) consumption of contaminated water and fish; 2) consumption of agricultural produce that may be irrigated with river water; 3) exposure to contaminated water and bottom sediments during recreational and industrial use of the water; and 4) exposure to build-up of sludge and deposits in water systems utilizing river water. The major sources of exposure are currently the ingestion of contaminated water and fish.

Mathematical models were developed for internal dose calculations and include the differences in intake and in size of the critical organ as a function of the individual's age. It was found that the skeleton of man received the largest exposure and ^{90}Sr was responsible for more than 99% of the dose equivalent. Based upon realistic but conservative assumptions, the estimated total doses from internal and external sources (20 year period) received by the skeleton of critical population groups, the 18-year-old individuals (in 1944) utilizing the Clinch River and the 14-year-old individuals (in 1944) utilizing the Tennessee River were 3.2 rem and 0.45 rem, respectively. These values are about a factor of 10 less than permissible limits established by ICRP and FRC.

Methods of calculation are presented, and examples are given for the transfer of ^{90}Sr and ^{137}Cs to man by irrigation water. If irrigation is practiced on the Clinch River in the future, it may become the dominant exposure pathway. It is noteworthy that in 1962 and 1963 fallout from nuclear tests contributed the same quantity of the critical radionuclide, ^{90}Sr to the Clinch River as did purposeful releases from the Laboratory.

INTRODUCTION

When radioactive material is released to a body of water, there is a complex network of mechanisms by which the material can be transmitted from one component, animate or inanimate, to another. At each point in the network or chain of transmission, human or other life forms may be subject to some degrees of radiation exposure.

The probability of human exposure and the degree of exposure depend upon many interrelated factors. These include: (1) the adequacy of control measures to keep the levels of contamination within safe limits; (2) the sources, types, quantities, and distribution of radioactive contaminants released to the water; (3) the physical, chemical and biological conditions in the body of water; (4) the use of water for drinking, domestic, and industrial purposes; and (5) the number of people exposed and their habits which may influence the nature and extent of exposure. Definitive information about these and other pertinent factors is necessary for realistic estimates of the potential exposures and evaluation of their significance.

In the Clinch River Study, safety evaluation depends primarily upon descriptive and analytical data needed to define exposure factors. Criteria of permissible radiation exposures, adopted by the International Commission on Radiological Protection (ICRP), the Federal Radiation Council (FRC), and the National Committee on Radiation Protection and Measurements (NCRP), are accepted as guides.[1-5] The MPC_w values employed to assess radiation dose to man follow the recommendations of ICRP found in Publication 2 and Publication 6. In Publication 6 the ICRP recommended an increase in the MPC_w values of ^{90}Sr when the skeleton and total body are the organs of reference, and they are used accordingly. On these bases, estimates of human exposure that may result from Clinch-Tennessee River contamination are made and conclusions reached regarding their importance.

Objectives of Study

The immediate objective is to evaluate the potential contribution of each relevant pathway in causing radiation exposure to man. The most direct means of evaluating internal exposures is to determine the amounts of radioactive material in the bodies of exposed members of population groups; for example, by whole-body counting or excretion analyses. The quantity of ^{137}Cs in the total body of eleven employees of the Oak Ridge Gaseous Diffusion Plant was measured by whole-body counting.[6] All were known to drink treated water from the Clinch River during working hours. Results were inconclusive, however, because the amount of ^{137}Cs in other parts of the diet due to fallout from weapons tests precluded an estimate of the proportion of the measured body burden that was attributable to consumption of Clinch River water. Therefore, exposures were calculated from measurements of the amounts of radioactive material in the various environmental media, with assumptions as to the fraction of this material that may affect the exposed population.

The long-range objectives are evaluation of the total potential of radioactivity in this river environment in causing exposures and delineation of exposure pathways so as to estimate the prevailing levels of safety and understand the potential for exposure of each such pathway in the future. The study is also directed toward establishment of parameters that affect downstream exposure from river disposal under many combinations of conditions.

Limitations of Analysis

Although human or other life forms may receive some degree of radiation exposure, this study does not consider effects upon biota in general but rather confines its efforts to estimation of radiation doses to man. The critical population groups may be identified from information about the critical radionuclides and principal exposure pathways, and with knowledge of the population distribution and habits. Not all of the desired information is available. For example, in order to complete some calculations, it is necessary to estimate the dietary habits and amounts of principal food-stuffs consumed as well as occupational and recreational habits. It is also desirable to confirm several estimates of external radiation which were calculated from measured concentrations of radionuclides in environmental media.

RADIONUCLIDES RELEASED AND CONCENTRATIONS IN THE RIVERS

Virtually all radioactive materials emanating from the Laboratory and reaching the Clinch River passes through White Oak Creek. The final control point for waste water released to the river at Clinch River Mile (CRM) 20.8 is at White Oak Dam(Fig. 1).

Discharges from White Oak Creek to Clinch River

The flow of water through White Oak Dam has been determined by several methods.[7,8] During the period 1953-1955, while White Oak Lake was still impounded, a gaging station at the dam was used. After 1955, when the lake was drained and the gaging station inactivated, flow was calculated by summing the separate measurements of flow in White Oak Creek and Melton Branch which are the principal surface streams draining the basin. The gaging station at the dam was reactivated in 1960 and has been used for flow measurements since that time.

All liquid waste handling systems, points of effluent release, and surface waters within ORNL are extensively monitored and sampled. Continuous proportional samples are collected of all process waste released to White Oak Creek and of all effluents released from White Oak Dam to the Clinch River. These samples are analyzed every 24 hours for at least gross beta activity and daily or weekly for gross alpha activity. The equipment employed in the routine analysis is capable of detecting beta particles with energies at least as low as 0.1 Mev. Continuous monitors are in operation on the Process Waste System and in White Oak Creek and White Oak Dam that are capable of detecting the beta particles (0.22 Mev) emitted by ^{147}Pm. This system of monitoring and sampling can be expected to alert Laboratory personnel to any unusual releases of radionuclides not determined in monthly composite sampling; that is, beta emitters that are shorter lived and less energetic than those normally encountered.

Until 1948, daily radiation measurements were made at White Oak Dam. Samples were collected periodically and analyzed for gross-beta activity. The number of beta curies released was calculated using mean annual discharges (daily flow measurements) and either measured or estimated gross activity content as follows: 1944, 600 curies; 1945, 500 curies; 1946, 900 curies; 1947, 200 curies; and 1948, 496 curies. Only infrequent

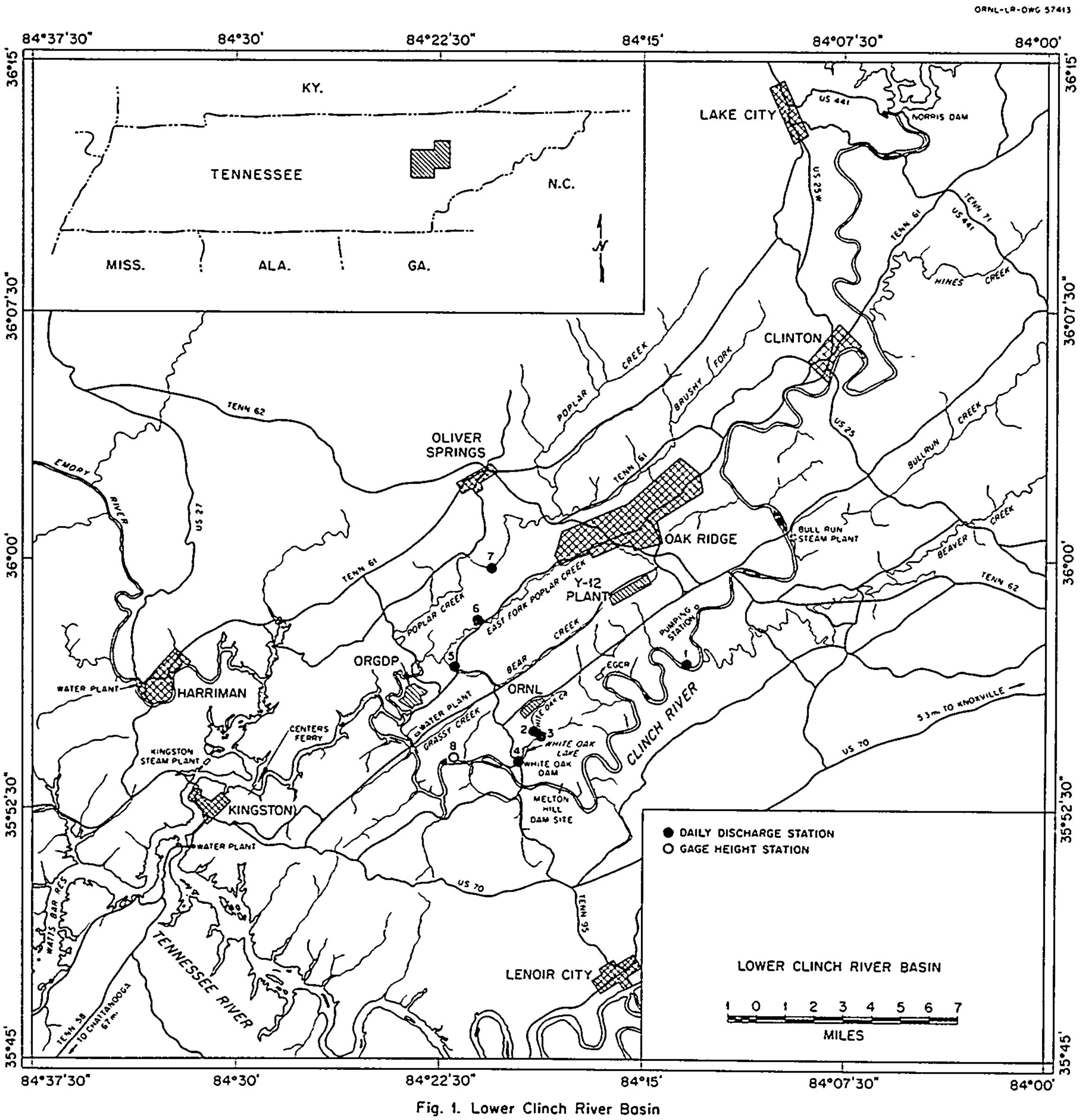


Fig. 1. Lower Clinch River Basin

Fig. 1. Lower Clinch River Basin

radiochemical analyses were made. An estimate was made of the ^{90}Sr content from the average amount, 27% found to be present during the period 1949 to 1958. For the purpose of further calculations, ^{90}Sr released from 1944 to 1948 was estimated as follows: 1944, 150 curies; 1945, 120 curies; 1946, 240 curies; 1947, 60 curies; and 1948, 130 curies.

Beginning in 1949 monthly composite samples were also analyzed radiochemically for cesium, ruthenium, strontium, cobalt, trivalent rare earths (TRE), cerium, zirconium, niobium, and iodine; and the curies released each year were calculated (Table I). The increased quantity of ^{137}Cs released in 1955 was due to the draining of White Oak Lake. Subsequent reduction in release of this nuclide was associated with treatment of process waste water and partial reimpoundment of the lake. The increase in ^{106}Ru released was associated with operation of the waste pits, while the decrease in ^{90}Sr released was related to the operation of the Process Waste Water Treatment Plant and modified waste management practice. It is noteworthy that the quantity of ^{90}Sr released to the Clinch River in 1962 and 1963 was about the same as that contributed by fallout from weapons tests.

Concentrations and Potential Human Exposures Downstream

Estimates of the mean annual concentrations of radionuclides in the Clinch and Tennessee Rivers were based on dilution ratios and the fact that White Oak Creek effluent is completely mixed with river water after 3 to 5 miles of flow downstream from the mouth of the creek. This was shown by tracer tests in the Clinch River in 1958, 1961, and 1962.[9,10,11] The concentration values derived in this way are conservative, since no allowance was made for decreases of the radionuclides in the water by radioactive decay or removal with suspended sediments.

Four downstream locations were considered for the evaluation analyses (Fig. 1,2), namely: (1) Clinch River Mile (CRM) 14.5, which is 6.3 miles downstream from the White Oak Creek discharge at CRM 20.8; (2) CRM 2.6, downstream from the mouth of Emory River near Kingston Steam Plant; (3) Tennessee River Mile (TRM) 529.9, Watts Bar Dam and Resort water supply; and (4) TRM 465, at Chattanooga water supply intake and 6.0 miles downstream from Chickamauga Dam.

TABLE I

YEARLY DISCHARGES OF RADIONUCLIDES TO CLINCH RIVER (CURIES)[a]

Year	Gross Beta	^{137}Cs	^{106}Ru	^{90}Sr	TRE(-Ce)	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co
1949	718	77	110	150	77	18	180	22	77	
1950	191	19	23	38	30		15	42	19	
1951	101	20	18	29	11		4.5	2.2	18	
1952	214	9.9	15	72	26	23	19	18	20	
1953	304	6.4	26	130	110	6.7	7.6	3.6	2.1	
1954	384	22	11	140	160	24	14	9.2	3.5	
1955	437	63	31	93	150	85	5.2	5.7	7.0	6.6
1956	582	170	29	100	140	59	12	15	3.5	46
1957	397	89	60	83	110	13	23	7.1	1.2	4.8
1958	544	55	42	150	240	30	6.0	6.0	8.2	8.7
1959	937	76	520	60	94	48	27	30	0.5	77
1960	2190	31	1900	28	48	27	38	45	5.3	72
1961	2230	15	2000	22	24	4.2	20	70	3.7	31
1962	1440	5.6	1400	9.4	11	1.2	2.2	7.7	0.36	14
1963	470	3.5	430	7.8	9.4	1.5	0.34	0.71	0.44	14

[a]Values calculated from data supplied by Applied Health Physics Section, ORNL.

Information regarding community water systems on or near the Clinch and Tennessee Rivers downstream from ORNL to South Pittsburgh, Tennessee, is given in Table 2. At CRM 14.5 and on the Emory River in the vicinity of CRM 4.4, water supplies taken from the river are used for sanitary and industrial purposes by the Oak Ridge Gaseous Diffusion Plant and Kingston Steam Plant, respectively. There are downstream recreational areas at the Kingston waterfront, at Watts Bar Dam, and at numerous places along Watts Bar Reservoir. Also, there are large recreational areas along Chickamauga reservoir, notably just above Chickamauga Dam (CRM 471.0). The first large population center (Chattanooga, Tennessee) is located a few miles downstream from Chickamauga Dam (TRM 471.0) and is served by a public water supply taken from the Tennessee River at TRM 465. In addition, CRM 14.5, TRM 529.9, and TRM 471.0 are stations in the basic water-sampling network of the Clinch River Study. The quantity of water passing each location annually was calculated from average flow values (Table 3). The average concentration of radionuclides at each location was determined from the curies released and the total flow for each year.

In this report the calculated concentration values for two of the locations are given, namely: CRM 14.5 and TRM 465 (Tables 4 and 5). Analyses for ^{91}Y were not performed. For the purpose of estimating dose, the concentrations of ^{91}Y were assumed to be equal to the difference in the concentration of trivalent rare earths and the concentration of ^{90}Sr (in equilibrium with ^{90}Y).

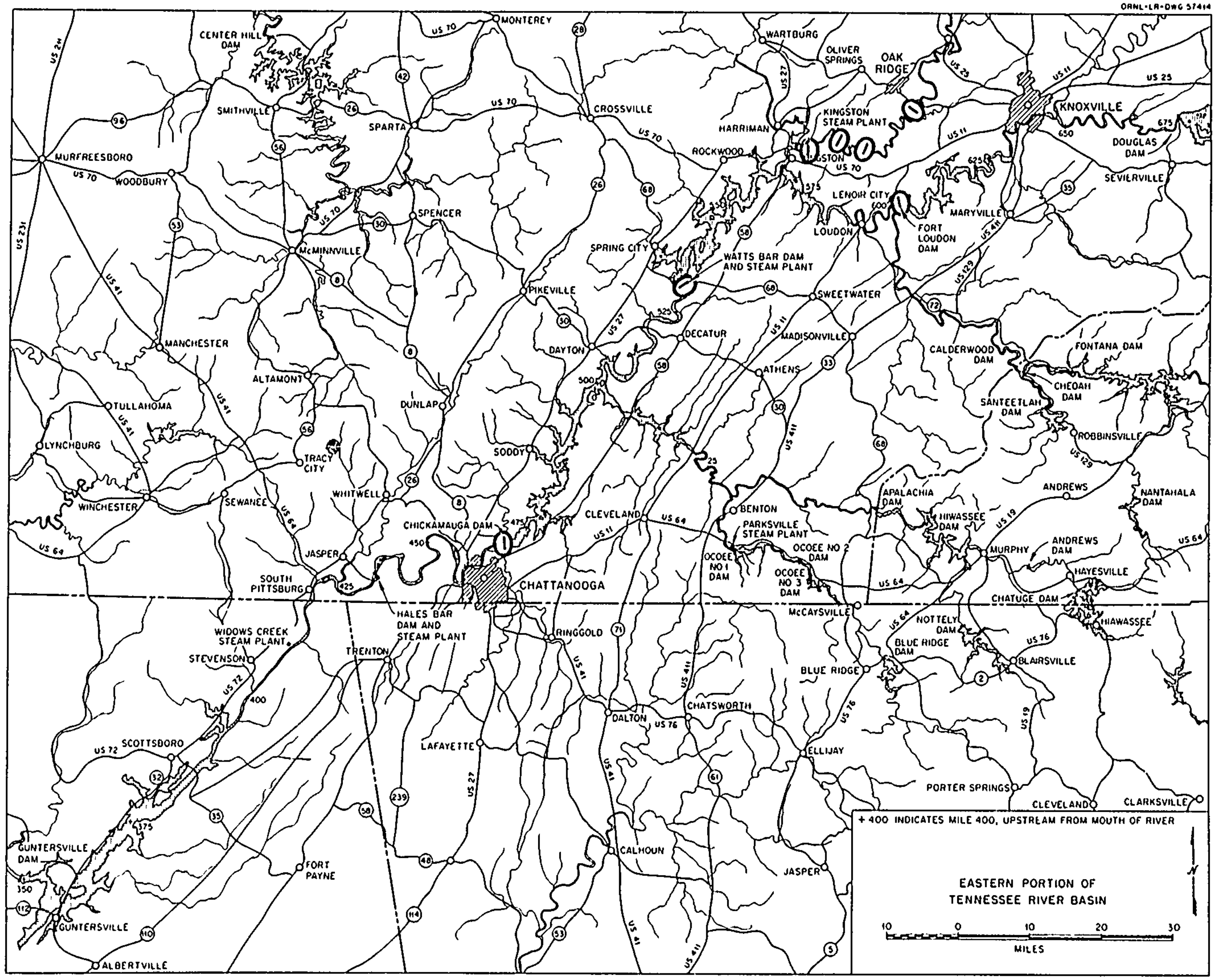


Fig. 2. Network Water Sampling Stations.

TABLE 2

COMMUNITY WATER SYSTEMS IN TENNESSEE DOWNSTREAM FROM ORNL SUPPLIED BY INTAKES ON CLINCH AND TENNESSEE RIVERS OR TRIBUTARIES THAT MAY BE AFFECTED BY MAIN STREAM CONDITIONS

Community	Intake Source		Number of Services	Population Served	Quantity (MGD)	Remarks
	Stream	Location				
ORGDP K-25	Clinch R.	CRM 14.5		3,015	4	Industrial plant potable water system.
Harriman	Emory R.	ERM 12	2,858	12,000	1.15	May at times draw Clinch R. water.
Kingston Steam Plant	Emory R.	CRM 4.4		600	.05	Potable water system.
Kingston	Tenn. R.	TRM 568.1	1,265	6,500	.29	River supplements spring supply.
Rockwood	Tenn. R.	TRM 553	2,000	7,000	1.0	River supplements spring supply.
Spring City	Piney R.	PRM 6.4	611	1,850	.15	Piney R. supplements spring supply.
Watts Bar Dam and Resort	Tenn. R.	TRM 530	25	150	.03 .14	Summer population highly variable.
Soddy-Daisy-Falling Water Walden's Ridge	Soddy Creek Embayment	TRM 488	2,545	8,000	.4	Supply approximately 3/4 from river, 1/4 from well.
Harrison Bay State Park	Tenn. R.	TRM 478		50	.05	Population highly variable. Swimming pool separate.
Booker T. Washington State Park	Tenn. R.	TRM 474			.05	Supplies swimming pool only.
Volunteer Ordnance Works-Farmers Chemical Association	Tenn. R.	TRM 473		300	37	Water used in processing.
Chattanooga	Tenn. R.	TRM 465	50,000	225,000	38.0	Includes Signal Mtn.
South Pittsburg	Tenn. R.	TRM 435	1,300	4,000	.4	

TABLE 3

MEAN ANNUAL FLOW IN CLINCH AND TENNESSEE RIVERS
(CUBIC FEET PER SECOND)

YEAR	CRM 14.5[a]	CRM 2.6[a]	TRM 529.9[b]	TRM 471.0[b]
1944	4800	6870	25690	32290
1945	4940	7020	26490	32270
1946	5150	6880	29100	38540
1947	4420	5720	24040	31190
1948	4290	6480	26370	34360
1949	5460	7560	33300	43630
1950	6630	9360	34240	44030
1951	6170	8760	28070	36560
1952	4570	5770	22470	29770
1953	4340	5710	22160	28130
1954	2990	4730	20480	26050
1955	4850	6610	23790	30530
1956	5040	7340	24750	30990
1957	6350	9300	36310	45250
1958	5560	6880	27780	34330
1959	3490	5260	23760	29000
1960	4460	6200	25150	31010
1961	4780	7110	29520	37430
1962	4980	8400	33700	40600
1963	5110	7180	25400	31600

[a]Values furnished by the United States Geological Survey - Estimated on basis of discharge records for the gaging station on Clinch River near Scarboro and intervening inflow.

[b]Values furnished by the Tennessee Valley Authority.

TABLE 4

CALCULATED MEAN ANNUAL CONCENTRATION OF RADIONUCLIDES

AT CLINCH RIVER MI. 14.5

(UNITS OF 10^{-9} $\mu c/ml$ or pc/liter)

Year	Gross Beta	^{137}Cs	^{106}Ru	^{90}Sr	^{91}Y	^{144}Ce	^{95}Zr	^{95}NB	^{131}I	^{60}Co
1944	100			40						
1945	100			30						
1946	200			50						
1947	60			20						
1948	130			40						
1949	150	16	22	30	0	3.7	36	4.6	16	
1950	32	3.2	3.9	6.5	0		2.5	7.2	3.2	
1951	18	3.6	3.2	5.2	0		0.82	0.40	3.2	
1952	53	2.4	3.6	18	0	5.6	4.7	4.4	4.8	
1953	78	1.7	6.8	35	0	1.7	2.0	0.93	0.54	
1954	140	8.2	4.2	51	11	8.9	5.2	3.5	1.3	
1955	100	15	7.1	22	13	20	1.2	1.2	1.6	1.5
1956	130	38	6.5	23	7.6	13	2.6	3.4	0.78	10
1957	70	16	11	15	5.5	2.2	4.0	1.3	0.21	0.85
1958	110	11	8.4	30	18	6.0	1.2	1.2	1.7	1.8
1959	300	25	170	19	11	16	8.7	9.5	0.16	24
1960	550	7.7	480	6.9	5.1	6.7	9.3	11	1.3	18
1961	520	3.5	480	5.2	0.35	0.98	4.6	17	0.87	7.3
1962	270	1.0	260	1.8	0.30	0.23	0.40	1.4	0.067	2.6
1963	100	0.76	94	1.7	0.35	0.33	0.074	0.16	0.096	3.1

TABLE 5

CALCULATED MEAN ANNUAL CONCENTRATION OF RADIONUCLIDES

AT TENNESSEE RIVER MI. 465

(UNITS OF 10^{-9} $\mu c/ml$ or pc/liter)

Year	Gross Beta	^{137}Cs	^{106}Ru	^{90}Sr	^{91}Y	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co
1944	20			5						
1945	20			4						
1946	20			6						
1947	8			2						
1948	16			4						
1949	18	2.0	2.7	3.7	0	0.46	4.5	0.57	2.0	
1950	4.9	0.49	0.60	0.98	0		0.38	0.11	0.49	
1951	3.1	0.61	0.54	0.87	0		0.14	0.068	0.54	
1952	8.1	0.37	0.56	2.7	0	0.86	0.72	0.68	0.74	
1953	12	0.26	1.1	5.4	0	0.27	0.30	0.14	0.083	
1954	17	0.94	0.48	5.8	1.2	1.0	0.59	0.40	0.15	
1955	16	2.3	1.1	3.4	2.0	3.1	0.19	0.21	0.26	0.24
1956	21	6.2	1.1	3.8	1.2	2.1	0.42	0.55	0.13	1.7
1957	9.8	2.2	1.5	2.1	0.78	0.31	0.56	0.18	0.030	0.12
1958	18	1.8	1.4	4.8	3.0	0.97	0.20	0.20	0.32	0.28
1959	36	2.9	20	2.3	1.3	1.9	1.1	1.1	0.02	2.8
1960	79	1.1	67	1.0	0.73	0.96	1.3	1.6	0.19	2.6
1961	67	0.47	61	0.67	0.05	0.13	0.59	2.1	0.11	0.93
1962	40	0.15	38	0.26	0.044	0.034	0.059	0.21	0.010	0.38
1963	17	0.12	15	0.28	0.056	0.053	0.012	0.025	0.016	0.49

AVENUES OF HUMAN EXPOSURE

Mechanisms of Exposure

The potential avenues of human exposure resulting from release of radioactivity to the environment are many and complex. H. M. Parker has indicated a number of exposure pathways and has suggested those which he believed to be of major consequence.[12] From radioactive wastes in rivers, streams, lakes, or reservoirs, he emphasizes the hazards related to use as drinking water, immersion in the water, close approach to the water (including contaminated mud and vegatation), use of water for irrigation, uptake by biological chains, industrial processes, sewage disposal, and atmospheric discharges.

The list is well conceived but, unfortunately, includes many avenues for which data are not available. An estimate of total human radiation exposure through surface waters is not possible now, and probably will not be for many years to come. However, based on available experience, the avenues of human exposure considered in the present report are believed to include the significant or potentially significant mechanisms of exposure resulting from radionuclide discharge to the Clinch River.

Critical Organs Considered

For a detailed estimate of exposure to radioactive material in the environment, it is necessary to calculate the dose to those organs for which the dose may reasonably be expected to be a maximum or to be in excess of the prescribed limits. To reduce the number of calculations, an insight concerning the potentially critical organs may be obtained by considering the type and concentration of radionuclides released, the maximum permissible concentration in water (MPC_w) for these radionuclides, the potentially significant avenues of exposures, and the type of individual under consideration. Based upon these considerations, the organs selected for analyses in this report include bone, gastrointestinal tract, thyroid, gonads, and total body. The bone and total body are reasonable selections when ^{90}Sr and ^{137}Cs are considered and when dose by immersion in contaminated fluids is possible. The increased quantity of

^{106}Ru, entering the surface water in 1960 and 1961, suggested analyses of the immersion dose and the GI tract. The genetic dose is of particular concern for exposure of a population and is included, although it can be estimated only approximately as equal to the total body dose; that is, equal to the average dose in other soft tissues. Finally, the release of ^{131}I implicates the thyroid, especially when the child is considered.

Estimation of Dosage to Organ

The fraction of MPC_w attained for the case of internal dose was calculated according to the recommendation of the ICRP.[1] For a mixture of invariant composition and based on a particular organ, x, the fraction of MPC_w that is attained is given by:

$$\sum_i \frac{P_{wi}}{(MPC)^x_{wi}} \qquad (1)$$

where

P_{wi} = the concentration of the particular radionuclide in water and

$(MPC)^x_{wi}$ = the maximum permissible concentration of the particular radionuclide in water for the organ and individual of interest and for continuous exposure.

When the value of expression (1) is less than or equal to 1, the exposure is not in excess of permitted limits. This formulation neglects the dose due to external sources, which will be estimated separately in this report.

The values of P_{wi} are to be average values, the period of averaging being one year according to the recommendations of ICRP, NCRP, and FRC (Table 6 lists the maximum permissible limits recommended by ICRP and FRC)[1,2,4] Thus transient changes in these environmental levels may not be of great significance. A high concentration in the river water on a given day is an important factor for operation of the facilities, and operating personnel will want to determine whether it is due to a change in the facilities or procedures, whether it results from reduced flow in the river (loss of dilution factor), et cetera.

TABLE 6

MAXIMUM PERMISSIBLE EXPOSURE

Agency	Type of Situation	Average Dose (rem/yr)			
		Bone[a]	G. I. Tract	Total Body	Thyroid
ICRP[c]	A. Occupational Worker	30	15	5	30
	B. Plant Vicinity				
	Work in vicinity[b]	3	1.5	1.5	3
	C. Population at Large				
	Individual	3	1.5	0.5	3
	Average	1	0.5	0.17[a]	1
FRC[c]	A. Occupational Worker	30	15	5	30
	B. Population at Large				
	1. Individual				
	a. Adult	1.5		0.5	3.0
	b. Child	1.5		0.5	1.5
	2. Population average				
	a. Adult	0.5		0.17	1.0
	b. Child	0.5		0.17	0.5

[a]Multiply by 0.3 to obtain portion of dose suggested for internal sources.
[b]Or visit area occasionally.
[c]See reference list No. 1,2,3.

However, if such a single measurement is used in formula (1), the result does not represent meaningfully the actual exposure. At best it represents only the hypothetical situation that would exist if the level persisted for at least a year. The fact that only values of the concentrations P_{wi}, averaged over a period of one year, are to be used in (1), is frequently overlooked. This has led, in some cases where transient levels have been high, to gross misinterpretations and unwarranted concern by the public.

Formula (1) is easily rearranged to represent a dose rate to organ x, but, again, the formula requires careful interpretation. If the exposure situation remains unchanged for 50 years, the weekly dose received by a particular organ due to internal and external sources is given by:

$$D_{50} = \sum_i {\frac{P_{wi}}{(MPC)_{wi}}}_x L + \sum_j R^x_j \qquad (2)$$

where

L = the average weekly dose (rem)* permitted to the organ, and

R_j = the weekly dose (rem) received by organ x from external sources of a particular radiation type

It is clear that the formula for D_{50} as given above only applies to a long-term and stable situation. The length of the period for application depends upon the effective half-life of the radionuclide involved. In the case of the Clinch River the presence of ^{90}Sr and other bone seekers as an important contributor to the dose means that the formula for D_{50} is directly applicable only for an exposure situation which is relatively stable over a long period of years. Thus the concentrations P_{wi} should be averages representative of the concentration in river water over long periods of years, and these concentrations are supposed to be constant

*The rem is defined by the International Commission on Radiological Units (ICRU) as the unit of dose equivalent. The dose equivalent is numerically equal to the dose in rads multiplied by the appropriate modifying factors.[13]

during that period. This greatly limits the direct usefulness of the formula D_{50}.

Dose Commitment for the Future

There is, however, a second interpretation of the formula giving D_{50} which is more useful in this situation. D_{50} can be interpreted as the dose that will be received during the next 50 years due to an exposure of one week with P_{wi} and R_j^x determined only for that week. With this interpretation D_{50} is a dose commitment for the future, at least in part, rather than a dose actually received during the week the individual was present in the area. Of course, the doses from external sources, that is, the dose represented by the term, $\sum_j R_j^x$, will be received during the period of occupancy of one week and not in any subsequent period.

The first terms of D_{50} represent the doses that will result from radionuclides entering the body during the period of occupancy of one week. The dose will be delivered during various periods following the intake, depending upon the effective half life of the radionuclide involved. For example, if the radionuclide in question is ^{131}I, the dose due to this intake would be received essentially during the following three or four weeks; but if the isotope in question is ^{90}Sr, then the dose would be distributed throughout the remaining 50 years of the person's life if he lives that long. In any case D_{50} gives the total dose commitment due to the individual's occupancy of the area during this week; that is, the dose which will be received during the next 50 years following the intake resulting from this occupancy.

Corrections for Dose Estimates Based on "Standard Man"

Even with this interpretation the formula D_{50} is subject to numerous reservations and requires further interpretation. Because the MPC's which enter into the formula have been estimated only on the basis of so-called "standard man," the dose represents only that which would be received by a person of physical characteristics and habits resembling standard man. Some examples of corrections that may be necessary for standard-man estimates are mentioned below.

Such estimates of dose rates or dose commitments should be considered as average values for typical adult individuals and considerable spread about these averages is to be expected. Both the FRC and the ICRP allow a factor of 3 as a practical range to provide for the variation of dose received in a homogeneous population group. This means that among adults, children of like age, and others with comparable characteristics it is assumed that only a small fraction will receive more than 3 times the average dose. The limited data available on actual exposures suggest that the dose received by only about 5% of an adult population would exceed this factor of 3 times the average.

The formula for D_{50} does not provide for any differences due to age, sex, or other variables that may affect the intake or metabolism of the radionuclide. Perhaps the most substantial correction is that required to take account of the child, the infant, or the fetus. During these early periods of life, the organs of the body are substantially smaller than those of standard man, and in some cases the intake and metabolism do not seem to differ to the same degree from those of standard man. Thus, a fairly large correction factor may be involved. Very little is known at the present time concerning differences in metabolic rates or processes of children and adults as they relate to important radionuclides. In this report it is not possible to make any adjustment on this basis. In the few cases where bits of data are available on children and infants, the difference of their metabolic rates from those of adults does not appear to be large. There remains, however, the difference due to intake and organ size. The charts shown in Fig. 3,4, and 5 have been prepared by M. J. Cook (of ORNL) to illustrate the magnitude of these differences as estimated on the basis of data at hand. The chart indicates a base line which represents the ratio of intake to organ weight for standard man. The curve represents the correction factor which adjusts for the change of this ratio with age. Assuming, as above, that metabolism is not substantially different for the infant or child, this graph gives a correction factor which can be applied to the dose estimate

$$\sum_i \quad P_{wi} L/(MPC)^x_{wi}$$

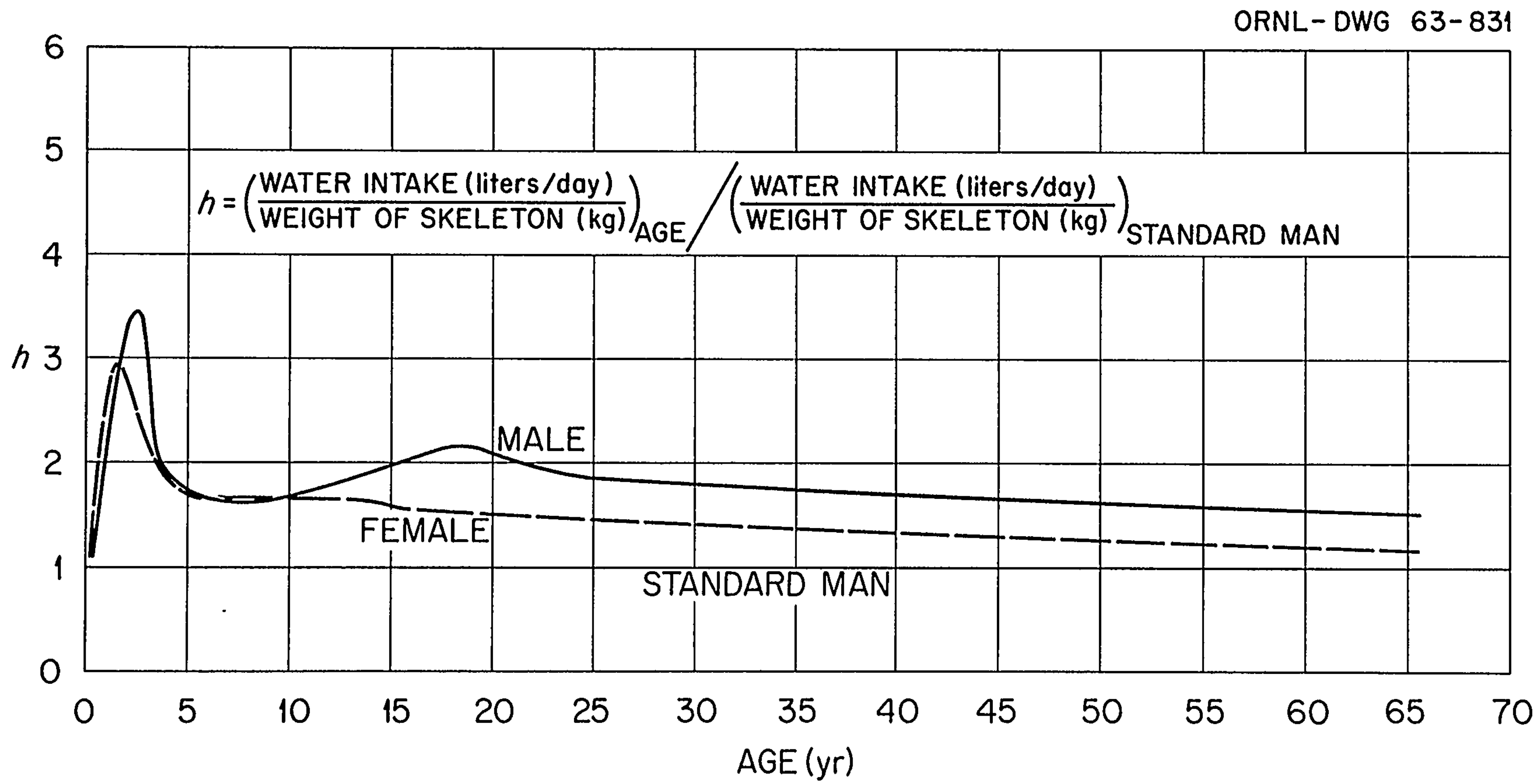


Fig. 3. Dose Correction Factor for Skeleton

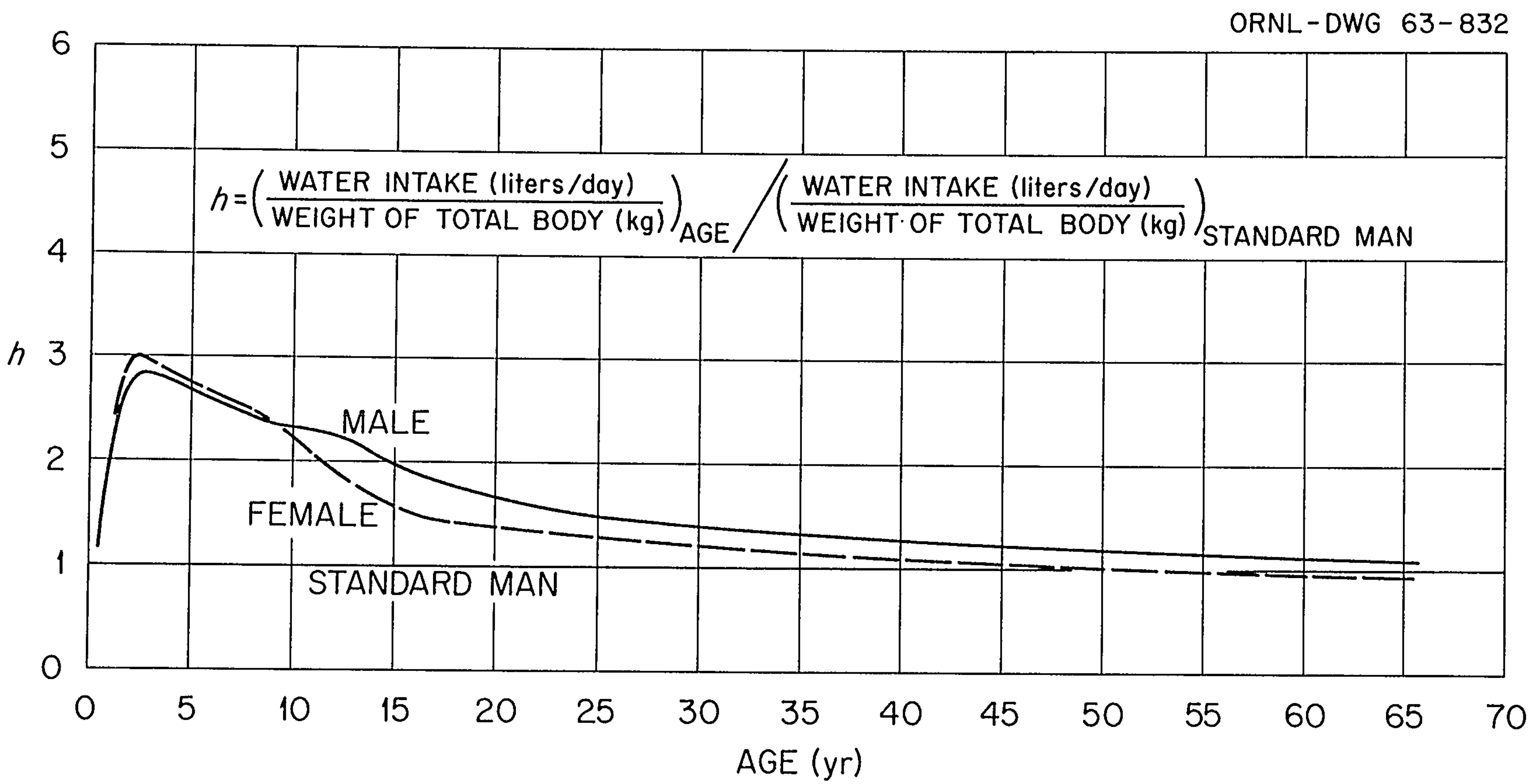


Fig. 4. Dose Correction Factor for Total Body.

ORNL-DWG 63-830

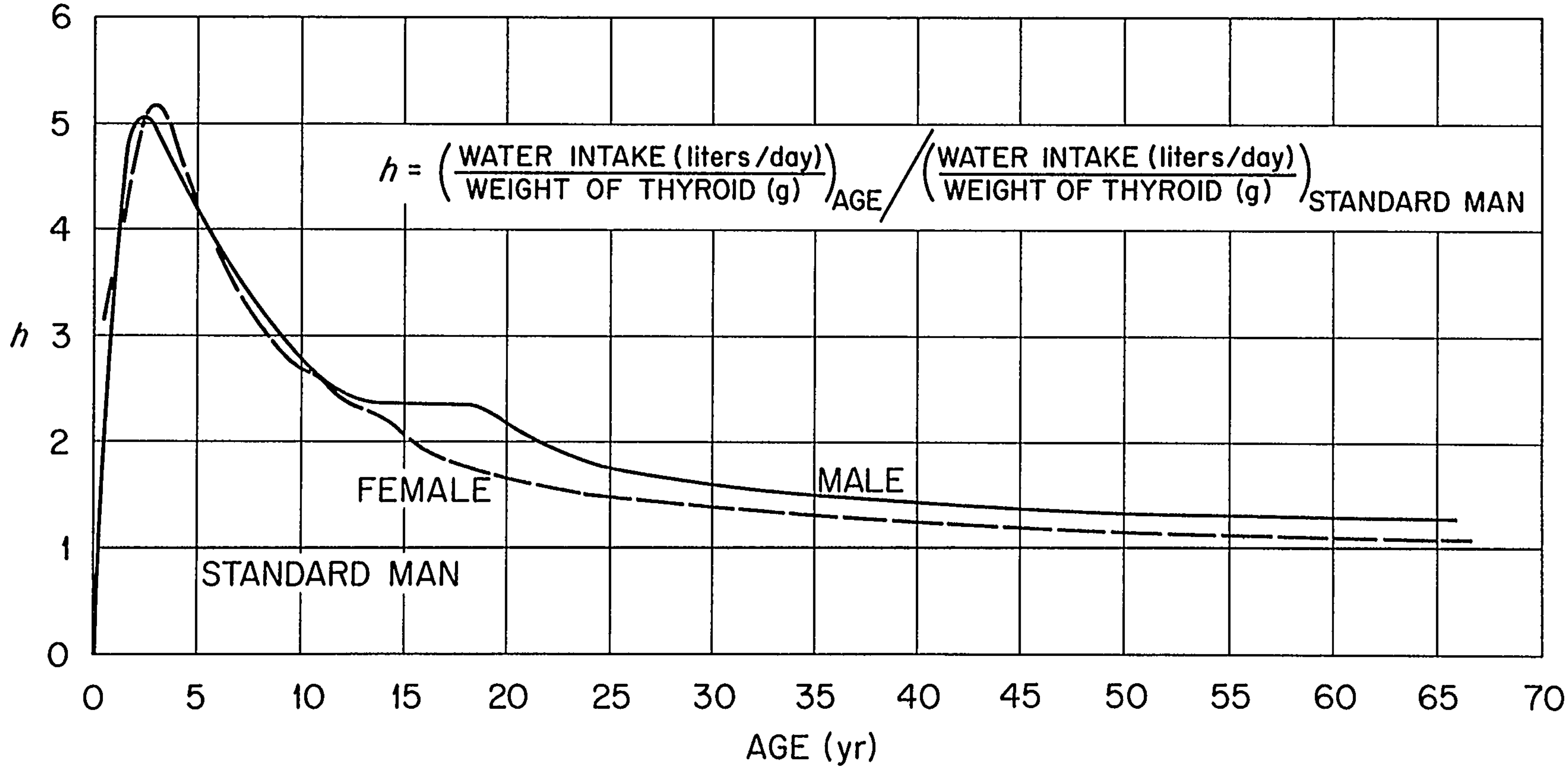


Fig. 5. Dose Correction Factor For Thyroid.

in the formula for D_{50} making it applicable for individuals of various ages. No very significant correction factor need be applied to the terms $\sum_j R_j^x$ so far as body size is concerned.

It is apparent from Figs. 3,4, and 5 that there is a significant correction to be made for infants and young children when dose to the skeleton, thyroid, or total body (genetic dose) is in question. The data on organ weights were taken from references supplied by M. J. Cook[14,15,16] and the data on water intake were obtained by the USPHS.[17]

In the case of the gastrointestinal tract, the calculation of the MPC is based on the assumption that the wall of the tract will receive 50% of the beta-gamma dose and 0.5% of the alpha particle dose delivered to the contents of the tract. To a very large extent this dose will be proportional to the concentration of the radionuclide in the contents of the tract. It will not vary greatly with the mass of the contents or with the diameter of the tract. Thus no very significant correction is necessary so far as the masses of the organ or contents are concerned. Assuming the tract is always full and that the residence time is short compared to the half-life of the radionuclides of interest, the dose received will not be changed significantly as residence time varies. This leaves the concentration of the radionuclide in the contents of the tract, and, hence, the dietary composition as the only variable of significance.

The ratio

$$\left(\frac{\text{Intake of Water}}{\text{Weight of Contents of GI Tract}}\right)_{\text{age}} \Bigg/ \left(\frac{\text{Intake of Water}}{\text{Weight of Contents of GI Tract}}\right)_{\substack{\text{standard}\\ \text{man}}}$$

would seem to be the appropriate correction factor to apply here. No data have been found on the variation of the weight of the contents of the GI tract with age.

Maximum Permissible Limits for Internal Exposures

Table 7 gives the fraction of $(MPC)_w$ of the river water calculated by using the average concentration of the various radionuclides for each year where such data were available. All $(MPC)_w$ values used for data relating

TABLE 7

FRACTION OF MPC IN WATER FROM CLINCH AND TENNESSEE RIVERS

Year	Clinch River Mi 14.5				Tennessee River Mi 465.5			
	Bone	G. I. Tract	Total Body	Thyroid	Bone	G. I. Tract	Total Body	Thyroid
1944	0.1		0.06	0.02	0.04		0.07	0.006
1945	0.08		0.04	0.01	0.03		0.06	0.005
1946	0.1		0.07	0.02	0.05		0.09	0.008
1947	0.05		0.03	0.01	0.02		0.03	0.003
1948	0.1		0.06	0.02	0.03		0.06	0.005
1949	0.076	0.0043	0.044	0.021	0.028	0.0016	0.054	0.0077
1950	0.016	0.0022	0.0094	0.0043	0.0073	0.0010	0.014	0.0021
1951	0.013	0.0017	0.0075	0.0038	0.0066	0.00087	0.013	0.0019
1952	0.044	0.0015	0.025	0.0098	0.020	0.00069	0.039	0.0045
1953	0.087	0.0018	0.050	0.015	0.040	0.00053	0.076	0.0053
1954	0.13	0.0032	0.072	0.022	0.044	0.0011	0.083	0.0074
1955	0.054	0.0037	0.032	0.0099	0.026	0.0019	0.050	0.0047
1956	0.059	0.0042	0.035	0.010	0.029	0.0020	0.057	0.0051
1957	0.037	0.0024	0.022	0.0063	0.016	0.00099	0.030	0.0027
1958	0.074	0.0031	0.043	0.013	0.034	0.0015	0.069	0.0077
1959	0.049	0.021	0.029	0.0084	0.018	0.0075	0.034	0.0030
1960	0.017	0.050	0.011	0.0037	0.0076	0.021	0.015	0.0016
1961	0.013	0.048	0.0077	0.0027	0.0050	0.019	0.0099	0.0010
1962	0.0044	0.026	0.0028	0.00083	0.0023	0.013	0.0040	0.00037
1963	0.0043	0.0096	0.0026	0.00093	0.0024	0.0052	0.0042	0.00038

TABLE 8

Maximum Permissible Concentrations of Radionuclides in Water[a]

(μc/ml)

Source of Supply	Nuclide	Critical Organ: Bone	Total Body	G. I. Tract	Thyroid
Clinch River	^{90}Sr	4×10^{-7}	7×10^{-7}	4×10^{-5}	2×10^{-6}
	^{89}Sr	1×10^{-5}	7×10^{-5}	3×10^{-5}	4×10^{-4}
	^{137}Cs	5×10^{-5}	2×10^{-5}	4×10^{-5}	1×10^{-4}
	^{106}Ru	1×10^{-3}	2×10^{-3}	1×10^{-5}	1×10^{-2}
	^{60}Co	6×10^{-4}	1×10^{-4}	3×10^{-5}	6×10^{-4}
	^{131}I	1×10^{-3}	2×10^{-4}	6×10^{-5}	2×10^{-6}
	^{95}Zr	2×10^{-1}	1×10^{-1}	6×10^{-5}	6×10^{-1}
	^{95}Nb	7×10^{-1}	4×10^{-1}	1×10^{-4}	2
	^{144}Ce	8×10^{-3}	3×10^{-2}	1×10^{-5}	2×10^{-1}
	^{91}Y	3×10^{-2}	2×10^{-1}	3×10^{-5}	1
Tennessee River	^{90}Sr	1×10^{-7}	7×10^{-8}	1×10^{-5}	8×10^{-7}
	^{89}Sr	3×10^{-6}	7×10^{-6}	1×10^{-5}	1×10^{-4}
	^{137}Cs	2×10^{-5}	2×10^{-6}	1×10^{-5}	4×10^{-5}
	^{106}Ru	3×10^{-4}	2×10^{-4}	3×10^{-6}	4×10^{-3}
	^{60}Co	2×10^{-4}	1×10^{-5}	1×10^{-5}	2×10^{-4}
	^{131}I	4×10^{-4}	2×10^{-4}	2×10^{-5}	7×10^{-7}
	^{95}Zr	7×10^{-2}	1×10^{-2}	2×10^{-5}	2×10^{-1}
	^{95}Nb	3×10^{-1}	4×10^{-2}	3×10^{-5}	8×10^{-1}
	^{144}Ce	3×10^{-3}	3×10^{-3}	3×10^{-6}	6×10^{-2}
	^{91}Y	1×10^{-2}	2×10^{-2}	1×10^{-5}	4×10^{-1}

[a]As recommended by ICRP (see Reference 1) values of MPC_W for continuous occupational exposure are reduced to 1/10 and applied to the Clinch River and are reduced to 1/30 for bone, thyroid, and G. I. tract as critical organ and to 1/100 for whole body as critical organ and applied to the Tennessee River. When the organ of reference is not listed in ICRP Publication 2 an independent estimate of the corresponding MPC_W value for continuous occupational exposure is obtained from the expression L^x (MPC)w TB/0.1, where L^x is the weekly dose rate permitted to organ x and $(MPC)_w TB$ is the maximum permissible concentration in water for total body.

to the Clinch River (see Table 8) are taken as 1/10 of the occupational $(MPC)_w$ values for exposure during the entire week (168 hours). To obtain $(MPC)_w$ values relating to the Tennessee River, the $(MPC)_w$ for continuous occupational exposure (168 hours/week) has been multiplied by 1/100 for whole body as critical organ and by 1/30 with thyroid, bone, and GI tract as the critical organs. These values are suggested by ICRP for application to exposure of persons living in the neighborhood of the plant, or for the average exposure of the population at large, respectively. If the fraction of $(MPC)_w$ given in Table 7 is multiplied by the appropriate dose rate from Table 6, an annual dose rate is obtained. However, it must be borne in mind that in the case of radionuclides of long effective half-life, this annual dose rate will be attained only if occupancy continues for many years. While the FRC has not extended recommendations to many of the radionuclides of interest here, it has recommended that Federal Agencies use the recommendations of the NCRP and the ICRP in such cases. In a few cases where intake values recommended by the FRC are available and differ from recommendations of the ICRP and NCRP, a slight adjustment of the present values will be necessary to obtain dose estimates by the procedure used by FRC.

To obtain values of dose commitment for children, an additional factor must be applied as indicated by Figs. 3, 4, 5. It must be realized that these values for a child only apply during a relatively short period of life. For ^{131}I the annual dose to an individual child's thyroid might be as high as 12 times the average dose to the thyroid of an adult, but this would be only during the first year or two of life, and even during these years most infants less than 2 years of age would only be at a level of 4 times the average adult value. For bone, the situation is more complicated. The factor of 4 applies only during the years of age from, say, 10 to 20, and it is unlikely that an equilibrium situation would be reached in the bone. Thus the annual doses to bone of an individual child during these years might be expected to be less than 12 times the average adult values. However, the dose commitment for the future would be increased by the full factor of 4 for the average teen-ager and by a factor of 12 for the higher group of teen-agers. This additional dose would be received over many subsequent years of the individual's life span.

Comparison of Internal Dose Recommendations of FRC and ICRP

The FRC has recommended a set of Radiation Protection Guides (RPG) applicable to normal peacetime operations. In Report No. 1, RPG values are given for occupational exposure as well as for exposure of the gonads or the total body in the case of population exposure.[2] These values are identical with those recommended by the ICRP.[18] In Report No. 2, specific guidance is given in connection with exposure of population groups to ^{226}Ra, ^{131}I, ^{90}Sr, and ^{89}Sr.[3] RPG values are listed for single-organ exposure of the thyroid, bone, and bone marrow.

For the case of the thyroid gland and ^{131}I, the FRC recommends a RPG value of 1.5 rem per year for individuals and 0.5 rem per year for the average of suitable samples of an exposed group.[3] These values are half the corresponding guides suggested by the ICRP for exposure of the population, since the suitable samples of FRC includes only children.[19] According to FRC, "...80 picocuries of ^{131}I per day would meet the RPG for thyroid for averages of suitable samples of an exposed population group of 0.5 rem per year." For adults, the RPG for the thyroid would not be exceeded by rates of intake higher by a factor of 10; that is, 800 picocuries per day. Based upon ICRP calculations, an MPC_w value for standard man that is equivalent to 0.5 rem per year is 3.3×10^{-7} μ c per milliliter; or a daily intake of about 730 picocuries.[1] Within the precision of the data employed by these agencies in arriving at these respective guides or limits this difference in rate of intake is not significant. Notice that the dose calculated by equations 8 and 10 apply to standard man and include a term, g_t, to account for the fraction attained of permissible intake. A dose-correction factor is then applied to these equations to account for differences in the intake and organ size of the individuals under consideration. The calculated doses to the thyroids of child and man due to ^{131}I are compatible with recommendations of both agencies even though differences in the radiosensitivity of the thyroid are not considered.

For the case of the bone and ^{90}Sr, FRC recommends an RPG value of 1.5 rem per year for individuals and 0.5 rem per year for averages of exposed populations.[3] No distinction is made between dose to the bone of children and adults. They consider that a continuous dietary intake of 600 picocuries per day would generally correspond to a bone dose of 0.5 rem per year to the average of suitable samples of an exposed population. The ICRP suggests that for somatic dose the average permissible level for large populations be one-thirtieth of the continuous occupational value; that is, about one rem per year to the bone. According to Publication 2 of ICRP the rate of intake of ^{90}Sr by standard man corresponding to a dose at equilibrium of 0.5 rem per year is 40 picocuries per day.[1] However, the MPC_w value and thus the rate of ^{90}Sr intake by standard man was changed in Publication 6 of ICRP.[5] They now consider that metabolic data provides a better estimate of MPC values for ^{90}Sr (bone as critical organ), than the single exponential model used previously. Although the MPC_w value was increased by a factor of four the permissible body burden and resultant dose to the bone remain unchanged. Thus the permissible intake of ^{90}Sr by standard man was increased by a factor of four and a daily intake of 160 picocuries now corresponds to a dose of 0.5 rem per year. At present the ICRP uses a relative damage factor of 5 for bone-seeking radionuclides other than radium. The maximum permissible body burden and the associated maximum permissible intake of ^{90}Sr is weighted by a relative damage factor of 5. Thus to compare the guides offered by FRC and ICRP it is necessary to multiply the daily intake of 160 picocuries of ICRP by a factor of 5. In view of the uncertainty concerning the body burden of ^{90}Sr and the effect associated with the corresponding dose to the bone of adults at equilibrium, the discrepancy in rates of intake (600 pc per day and 800 pc per day) is not considered significant. The rate of intake and resultant bone dose suggested by the two agencies are not compatible even though there is an apparent difference of two in the standard to be applied to the exposed population.

RADIATION EXPOSURES FROM ORDINARY USAGE OF THE RIVERS

Evaluation of Dosages from Drinking Water

Estimates of the fraction of maximum permissible dosages received from drinking Clinch River and Tennessee River water are based on concentrations of radionuclides in the raw water. This approach is conservative, because it assumes that there will be no reduction of radionuclides in the water by water treatment before drinking, and it makes no allowance for portions of the radionuclides that are in the bottom sediments which would not be expected to enter raw-water intakes. Future calculations may consider radionuclide removal by water plants and bottom sediments, but the data now available do not warrant it.

The fraction of MPC_w that would be attained by drinking water from the Clinch and Tennessee Rivers at the two reference stations, namely CRM 14.5 and TRM 465, have been given in Table 7. For the period 1943-1948, only estimates of ^{90}Sr concentrations are available. Thus the fraction of MPC_w attained for this period is based on the estimates of ^{90}Sr. Such calculations are warranted only because ^{90}Sr has been responsible for almost all the bone and total body dose as well as a significant part of the dose to the thyroid. Inherent in the calculation of these fractional values is the assumption that exposure is continuous for a period of 50 years to the mixture of radionuclides that is present during the particular year. For these mixtures of radionuclides in the raw water, estimated dose to the bone constitutes a greater fraction of the maximum permissible limit than does the estimated dose to the other body organs. This is attributable to ^{90}Sr released. The largest fraction of bone dose attained was 0.13 (13%) for the 1954 concentrations, assuming that the same concentrations continued for 50 years. For example, applying the most restrictive FRC limit of thyroid dose (for the average child of the population-at-large which is 1/60 of the continuous occupational exposure), the fraction of MPC_w that would be attained at CRM 14.5 during 1961 is less than 0.03 (3%). The increase in internal dose to the GI tract for 1960 and 1961 is due to the increased release of ^{106}Ru.

Estimation of Radiation Dose from Ingestion of Water

The MPC_w values are set by the requirement that in an environment where the level of contamination remains constant and the composition of the contaminants is unchanged, the dose rate (rems/week) for an adult after 50 years of exposure shall not exceed a recommended limit. During such a 50-year exposure period, equilibrium in the body is reached by most of the radionuclides, because their effective half life in the body is short compared to 50 years. However, in the case of ^{90}Sr, the allowable annual dose rate is reached only after 50 years of continuous exposure of an adult to the MPC_w. At earlier times the dose rate to the skeleton or total body of such an adult will be below the recommended limiting dose. For this reason, and also because the levels and composition of the contaminants are not constant, estimation of dose received by ingestion of known concentrations is desirable. A mathematical model has been developed that will allow calculation of dose received as a function of time.

Following ingestion of water, the activity present in a critical organ of the body at time t(after the start of ingestion) can be expressed as:[1,20]

$$Q = f_w \; S \; X \int_0^t e^{-\lambda_e t'} \, dt', \tag{3}$$

where

$Q = \mu c$ present in critical organ,

f_w = fraction of ingested radionuclide that is retained in the critical organ,

S = rate of intake of water, ml/yr

X = concentration of radionuclide in water during exposure, $\mu c/ml$

λ_e = effective decay constant of radionuclide, 1/yr, and

t' = a time variable.

Assuming that the concentration of a radionuclide in water is the average annual concentration and the rate of water intake is 2.2 liters/day (standard man), Eq. 3 is integrated over a time period of 1 year giving:

$$Q(1)_t = \frac{f_w\, S\, \overline{X}_t}{\lambda_e} \left[1 - e^{-\lambda_e} \right], \qquad (4)$$

where

$Q(1)_t$ = μc present in the critical organ at the end of t years due to the intake of water during that year, and

$\overline{X}_t$ = average annual concentration μc/ml of a radionuclide in water during a particular year, t.

After the exposure period t, the quantity of radionculide remaining in the critical organ is given by:

$$Q(A)_{t,\tau} = Q(1)_t e^{-\lambda_e \tau} \qquad (5)$$

where

τ = the years after a particular intake period, t, and $1 \le \tau \le n$. Since the quantity of water consumed by an individual is a function of the individual's age, the critical-organ burden is also a function of the individual's age. Thus, an intake correction factor j_γ (where γ is the individual's age during a particular intake period), must be applied to Eqs. (4) and (5). For example, assume that an individual of age $\gamma = 10$ began to consume contaminated water at the beginning of year, t = 1, the critical-organ burden of a particular radionuclide each year for a period of, say, 3 years would be determined as follows:

Period	Body Burden (μc)	
t = 1	$j_{10}\, Q(1)_1$	
t = 2	$j_{11}\, Q(1)_2 + j_{10}\, Q(A)_{1,1}$	(6)
t = 3	$j_{12}\, Q(1)_3 + j_{10}\, Q(A) + j_{11}\, Q(A)_{2,1}$	

The dose received by the critical organ during the period of intake, t, is

$$D(I) = \frac{MPD}{qf_2} \int_o^t Qdt', \tag{7}$$

where

MPD = the maximum permissible dose rate to a particular organ, rem/yr, and

$qf_2 = \frac{MPC_w\ S\ f_w}{\lambda_e} \left[1 - e^{-\lambda_e 50} \right]$, the fraction of radionuclide in the critical organ after 50 years of continuous exposure.

By substituting Eq. (3) in Eq. (7) and integrating over an exposure period of 1 year, the dose received by the critical organ during a particular exposure year, t, is

$$D(I)_t = \frac{MPD\ g_t}{1 - e^{-\lambda_e 50}} \left[1 - \frac{1 - e^{-\lambda_e}}{\lambda_e} \right], \tag{8}$$

where

$g_t = \frac{\bar{X}_t}{MPC_w}$, the fraction of MPC_w in water during a particular year, t.

After the exposure period t the critical organ will continue to be irradiated by the radionuclide retained from the exposure period (see page 17). The length of time for such residual exposure depends on the effective half life of the radionuclide. The dose after exposure is

$$D(A)_{t,\tau} = \frac{MPD\ g_t\ \lambda_e}{1 - e^{-\lambda_e 50}} \int_o^t e^{-\lambda_e t'} dt' \int_{\tau-1}^{\tau} e^{-\lambda_e t''} dt'', \tag{9}$$

where

t' and t'' = time variables.

Integration of Eq. (9) over an exposure period of 1 year and post exposure period, τ, gives

$$D(A)_{t,\tau} = \frac{MPD\ g_t \left[1 - e^{-\lambda_e}\right]}{\lambda_e \left[1 - e^{-\lambda_e 50}\right]} \left[e^{-\lambda_e(\tau-1)} - e^{-\lambda_e \tau}\right] \quad (10)$$

The total dose received by a particular critical organ to a particular radionuclide after a number of years of exposure is then the sum of Eqs. (8) and (10). A dose-correction factor must be applied to Eqs. (8) and (10) to account for differences in the intake and organ size of the individuals under consideration. The dose correction factor is

$$h_\gamma = \frac{S_\gamma / M_\gamma}{S_{sm} / M_{sm}}, \quad (11)$$

where

S_γ = the rate of water intake of an individual of age, γ

S_{sm} = the rate of water intake of standard man,

M_γ = the weight of the critical organ of an individual of age, γ, and

M_{sm} = the weight of the critical organ of standard man.

For an individual of age, $\gamma = 10$, who began to consume contaminated water at the beginning of year, $t = 1$, the dose received each year for a period of, say, 3 years would be determined as follows:

Period	Dose
$t = 1$	$h_{10}D(I)_1$
$t = 2$	$h_{11}D(I)_2 + h_{10}(M_{10}/M_{11})\ D(A)_{1,1}$ (12)
$t = 3$	$h_{12}D(I)_3 + h_{10}(M_{10}/M_{12})D(A)_{1,2} + h_{11}(M_{11}/M_{12})D(A)_{2,1}$

Computer Calculations of Internal Dose

The mathematical models (equations 4, 5, 8 and 10), with appropriate correction factors, are coded for Data Control 1604, permitting machine computations of critical organ burdens and dose received. Calculations are performed by assuming that individuals from birth through age 45 and standard man began in 1944 to drink untreated water from the Clinch River (mile 14.5) and from the Tennessee River (mile 465). They continue to drink water from these sources through 1963, following which water is obtained from an uncontaminated supply. It is also assumed that all water taken into the body in food or drink is equally contaminated and that intake and organ mass vary with age according to data in Figures 3, 4, and 5.

Comparative Examples of Computed Dose.- Examples of the computed annual dose received by the skeleton, total body, and thyroid of males drinking Clinch River water are shown in Figures 6, 7, and 8. At the end of 1963 the dose rate to the skeleton (Fig. 6) of the critical population group, the 14-year-old, is about twice that of standard man. The dose rate received by the male skeleton of other age groups is less than the 14-year-old (see Table 9). The differences in dose rate are attributed to differences in intake and size of the skeleton. Strontium-90 is responsible for more than 99% of the skeleton dose; thus, smaller releases of this radionuclide in 1950 to 1952 and 1960 to 1963 are reflected by a reduction in annual dose received by the skeleton. Notice that the maximum dose rate to the skeleton of the potentially critical group is considerably less than 1/10 of the permissible continuous occupational levels of ICRP (3.0 rem per year). Dose rate to the total body (Fig. 7) and thyroid (Fig. 8) of the critical groups in 1963 is about 50% greater than that of standard man. Dilution of wastes in the Tennessee River results in a reduced dose rate (by a factor of about 8) to the individual organs. The differences in dose rates received by the organs are similar to those found for the Clinch River (Table 9). In all cases the dose rates to the critical organ of the potentially critical groups is at least one order of magnitude below permissible levels. The dose rate received by males is greater than that received by females of all

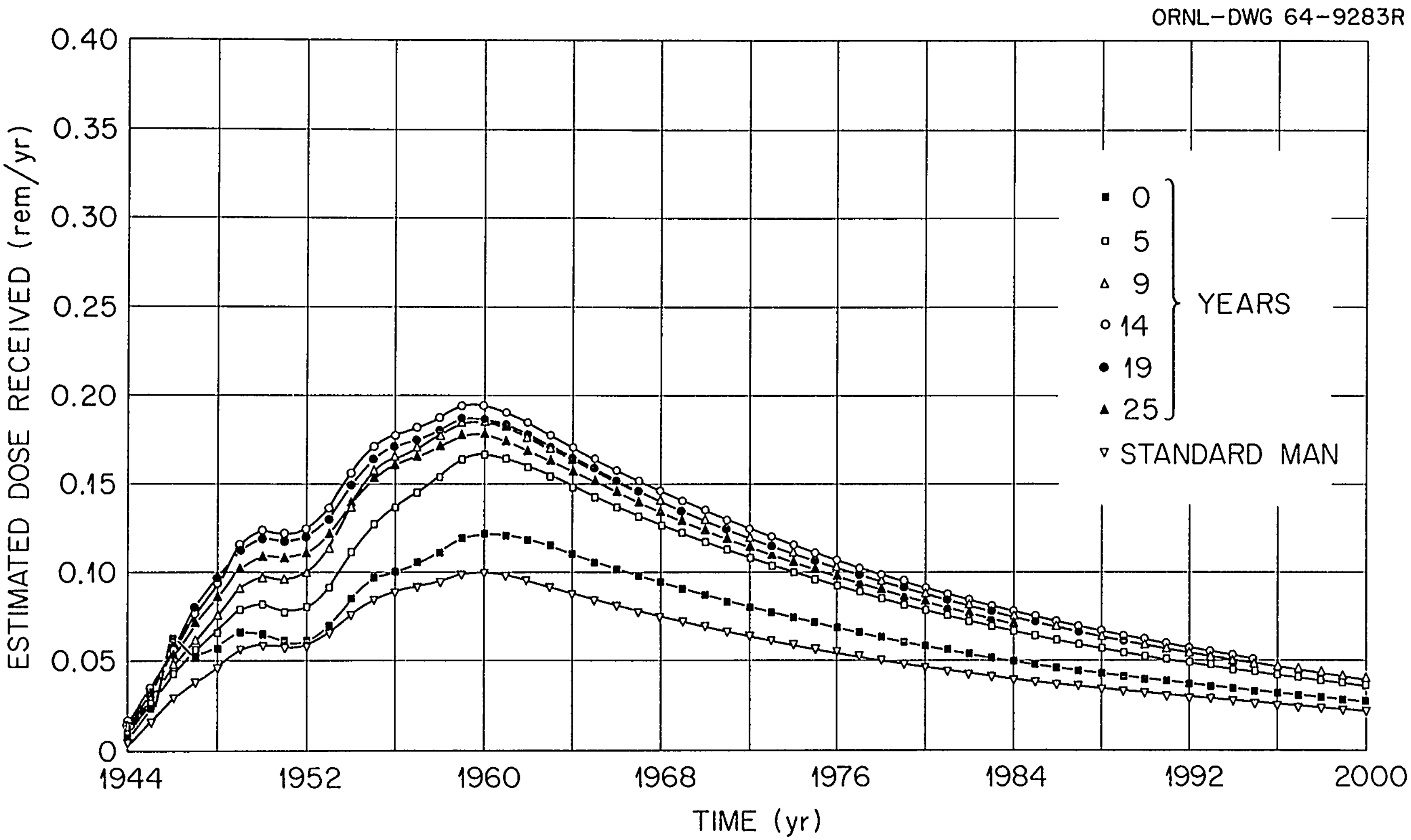


Fig. 6. Estimated Dose Received by Skeleton of Males from Drinking Clinch River Water.

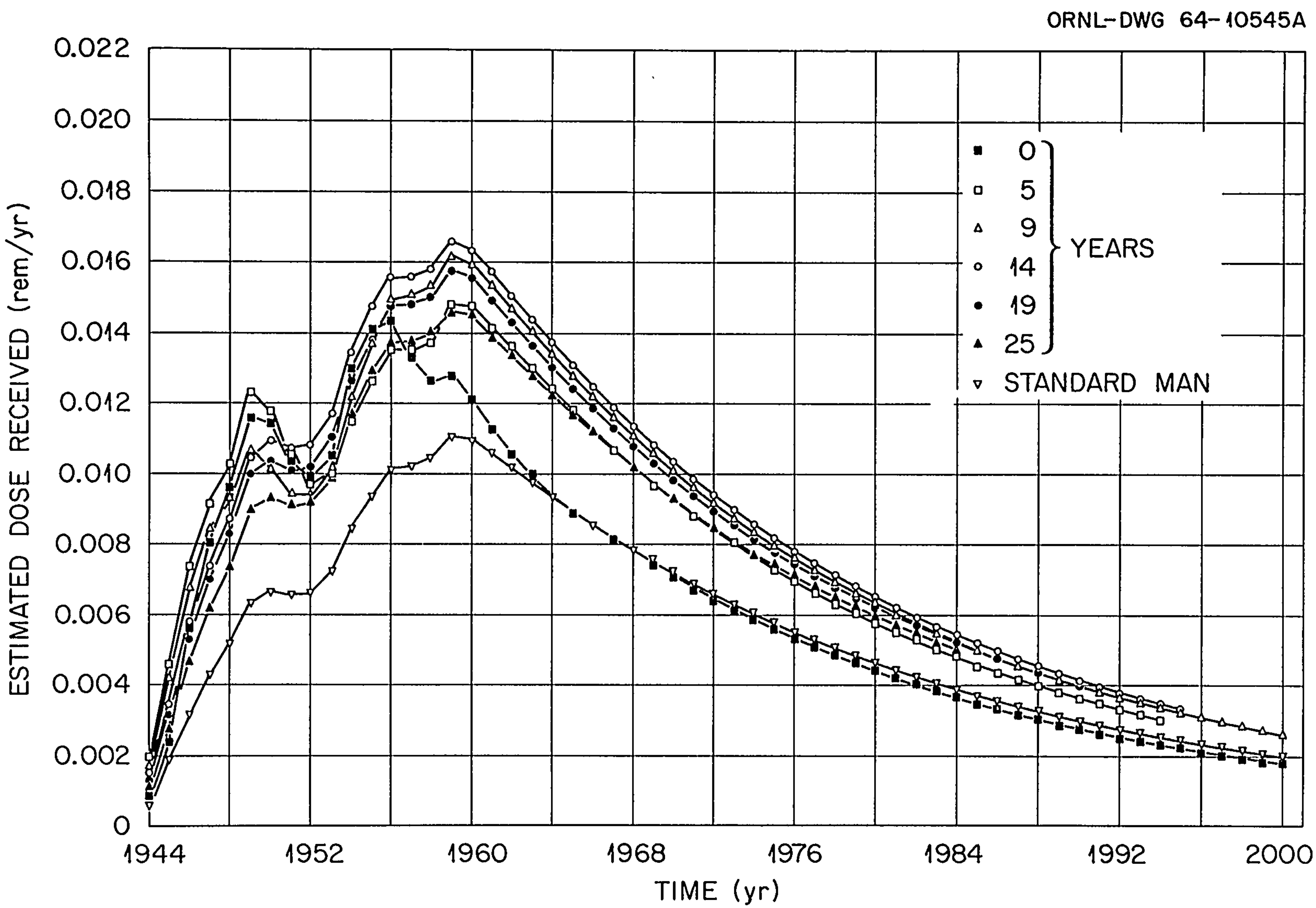


Fig. 7. Estimated Dose Received by Total Body of Males from Drinking Clinch River Water.

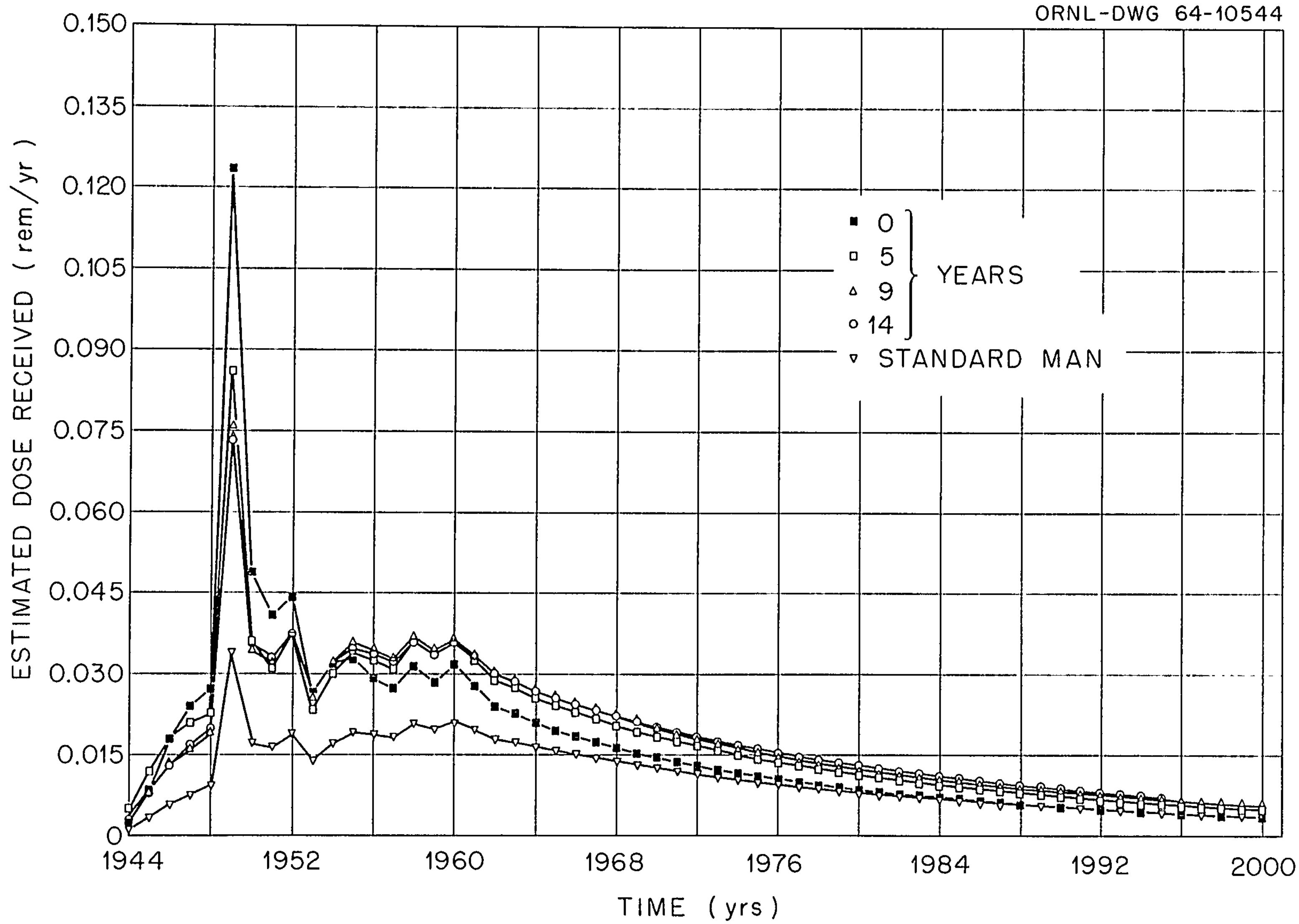


Fig. 8. Estimated Dose Received by Thyroid of Males from Drinking Clinch River Water.

TABLE 9

DOSE RATE TO CRITICAL ORGANS OF MALES FROM DRINKING WATER[a]

(rems/year)

Age at Start of Exposure	Clinch River Water			Tennessee River Water		
	Skeleton	Total Body	Thyroid	Skeleton	Total Body	Thyroid
0	0.16	0.0097	0.023	0.017	0.0014	0.0032
5	0.17	0.013	0.027	0.023	0.0018	0.0038
9	0.18	0.014	0.029	0.025	0.0019	0.0039
14	0.19	0.014	0.028	0.026	0.0019	0.0039
19	0.18	0.014	0.027	0.025	0.0018	0.0037
25	0.17	0.013	0.025	0.023	0.0017	0.0034
Standard Man	0.099	0.0097	0.017	0.013	0.0013	0.0024
Maximum Permissible Dose[b]	3.0	0.5	3.0	1.00	0.05	1.0

[a]The dose rate at the end of 1963.

[b]According to recommendations of the ICRP Publication 2 where values of annual dose rate for continuous occupational exposure are reduced to 1/10 and applied to the Clinch River and reduced to 1/30 for skeleton and thyroid as critical organ and to 1/100 for whole body as critical organ and applied to the Tennessee River.

age groups and critical organs considered.

Another interesting comparison is the total dose received by individuals during the period in which Clinch River and Tennessee River water is consumed. As shown in Table 10 the skeleton of a 14-year-old male receives a total dose of 2.9 rem by use of Clinch River water and 0.37 rem by use of Tennessee River water -- about twice that of standard man. About 99% of the total body dose is due to ^{90}Sr, and fluctuations in dose rate reflect changes in ^{90}Sr release as well as differences in intake and organ mass. The thyroid of the newborn infant receives the largest dose, about twice that of standard man. Strontium-90 and iodine-131 are responsible for 70% and 30% of the total thyroid dose, respectively. A large release of ^{131}I and the short effective half life of this radionuclide result in sizeable increase in thyroid dose during 1949. Of the organs analyzed, the skeleton of man receives the largest dose. After 1963 doses received by the critical organs is due to radionuclides that have accumulated during the period that contaminated water is consumed. This dose commitment should be considered in any assessment of future exposure likely to be received.

Consideration of Metabolic Factors

Recently information on metabolic processes of children and adults permits a preliminary assessment of their importance in estimating internal dose. In particular, Kulp and Rivera have examined the effects of bone growth, rate of bone turnover, and the ratio of strontium to calcium in bone to that in diet, on the retention of ^{90}Sr in the skeleton of man.[21,22] The difference equation for the model developed by Kulp (herein referred to as the age dependent metabolic model) can be expressed as:

$$S_n = S_{n-1} - (f+\lambda)\, S_{n-1} + K_n \frac{In\, C_n}{G_n} \left[f_n Ca_{n-1} + (Ca_n - Ca_{n-1}) \right] \quad (13)$$

where

S_n = pc ^{90}Sr skeleton burden at time n

f_n = fractional bone turnover rate during the period from time n-1 to time n

TABLE 10

DOSE RECEIVED BY CRITICAL ORGANS OF MALES FROM DRINKING WATER[a]
(rem)

Age at Start of Exposure (yrs after birth)	Clinch River Water			Tennessee River Water		
	Skeleton	Total Body	Thyroid	Skeleton	Total Body	Thyroid
0	1.7	0.20	0.65	0.23	0.026	0.087
5	2.3	0.22	0.61	0.30	0.029	0.082
9	2.6	0.23	0.60	0.34	0.029	0.079
14	2.9	0.23	0.59	0.38	0.030	0.078
19	2.8	0.22	0.53	0.36	0.028	0.070
25	2.6	0.20	0.48	0.34	0.026	0.063
Standard Man	1.5	0.15	0.32	0.19	0.019	0.042
Maximum Permissible Dose[b]	60	10	60	20	1	20

[a]The cumulative dose for the period 1944 to 1963.

[b]According to recommendations of the ICRP Publication 2 where values of annual dose rate for continuous occupational exposure are reduced to 1/10 and applied to the Clinch River and reduced to 1/30 for skeleton and thyroid as critical organ and to 1/100 for whole body as critical organ and applied to the Tennessee River.

λ = ^{90}Sr radiological decay constant per period

$K = \left(\frac{Sr}{Ca}\right) \text{Bone} \Big/ \left(\frac{Sr}{Ca}\right) \text{Diet}$

I_n = fluid intake, liters/day

C_n = ^{90}Sr concentration in water, pc/liter

G_n = calcium intake, grams/day, and

Ca_n = calcium content of the skeleton (grams).

The equation relates the loss and gain of ^{90}Sr during the period of interest, and is based on calcium metabolism. Calcium may enter and leave the skeleton each period. Bone turnover is defined as the quantity of calcium that enters and is excreted from the skeleton. The fractional bone turnover rate, F, is the bone turnover rate divided by the quantity of calcium contained in the skeleton. Strontium-90 in the skeleton is lost as a result of bone turnover and radiological decay and is gained by bone remodeling and bone growth.

By successive application of the difference equation, the skeletal burden can be determined. The dose rate is given by

$$Dn = a \frac{S_n}{W_n} \tag{14}$$

where

Dn = dose rate, rem/yr,

Wn = mass of the skeleton in kg at time n, and

a = a constant; for ^{90}Sr, 1.03×10^{-4} $\frac{\text{rems}}{\text{yr}} \Big/ \frac{\text{pc}}{\text{kg}}$.

These equations were coded for computer solution using an interval of one month during 0 to 2 years of age, three months during 2 to 24 years, and yearly intervals thereafter.

Estimation of the dose received by the skeleton of males due to ingestion of Clinch River water were performed using the same values for fluid intake, and ^{90}Sr concentration in water and skeleton mass, that were previously employed with the adjusted ICRP model in equations 8 and 10.[a]

[a]Calculations were performed by H. J. Fisher, U. S. Public Health Service, while working with the Internal Dose Estimation Section, Health Physics Division, ORNL.

Values of daily calcium intake and calcium content of the body were taken from Albritton and Mitchell, respectively;[14,23] values of bone turnover rate and the observed ratio (Kn) were from Rivera.[22] Examples of the cumulative skeletal dose (1944 to 1963) to individuals of various ages are given in Table 11. For comparative purposes, the table also includes the skeletal dose calculated with the adjusted ICRP model. In all cases the age dependent metabolic yields a slightly larger estimate of total dose (an average of 15% for the individuals listed) than the adjusted ICRP model. Unquestionably, changes can be expected in the values of metabolic factors as new information becomes available.

Dose Commitment Associated with Ingested Radionuclides

Dose commitments for the future (page 17) associated with the consumption of Clinch River and Tennessee River water are given in Table 12. These are estimated cumulative doses that persons of various ages receive, beginning in 1964 and extending to age 65; they result from the retention of radionuclides in critical organs due to ingestion of contaminated water during the period 1944 through 1963. For reasons previously enumerated, the critical organs of standard man have a smaller dose commitment (in general) than the organs of other individuals. In all cases the dose commitments are well below prescribed limits.

Exposures from Eating Contaminated Fish

Fish living in the Clinch River and Tennessee River downstream from White Oak Creek assimilate some of the radionuclides released to the river system. Since fish is a staple of man's diet, radionuclides present in the fish will contribute to his total dose.

Radionuclide Concentrations in Fish

The data on radionuclide concentrations in fish were obtained by the Subcommittee on Aquatic Biology, Clinch River Study Steering Committee.[24-29] Fish were collected during various seasons for the period of 1960 to 1962 and were processed to approximate, insofar as possible, normal human utilization.[25,29] Bottom feeders (carp, carpsucker, and buffalo) were processed either by grinding the flesh and bones together (total fish analyses) or by removing the flesh after cooking (flesh analyses). Sight

TABLE 11

DOSE RECEIVED BY SKELETON OF MALES FROM DRINKING WATER[a]

Age at Start of Exposure (yrs after birth)	(rem) Age Dependent Metabolic Model (Equations 13 and 14)	Adjusted ICRP Model (Equations 8 and 10)
0	2.2	1.7
5	2.4	2.3
9	3.1	2.6
14	3.3	2.9
19	3.3	2.8
25	2.9	2.6

[a]The cumulative dose from ingestion of Clinch River water during the period 1944 to 1963.

TABLE 12

DOSE COMMITMENT TO CRITICAL ORGANS OF MALES FROM DRINKING WATER[a]

(rem)

Age at Start	Clinch River Water			Tennessee River Water		
of Exposure	Skeleton	Total Body	Thyroid	Skeleton	Total Body	Thyroid
0	2.6	0.18	0.36	0.35	0.024	0.051
5	3.3	0.23	0.46	0.45	0.030	0.063
9	3.5	0.24	0.47	0.43	0.032	0.066
14	3.4	0.23	0.46	0.45	0.031	0.062
19	3.0	0.21	0.41	0.41	0.027	0.055
25	2.5	0.17	0.34	0.34	0.023	0.046
Standard Man	2.0	0.18	0.32	0.28	0.024	0.044

[a]The cumulative dose commitment beginning in 1964 and extending to age 65.

feeders (white crappie, bluegill, white bass, large mouth bass, sauger, and drum) were processed by removing the flesh after cooking. For the internal dose analysis, catfish were included with the sight feeders, since only the flesh of the catfish was processed. Another fish sampling program was completed May 1963. Carp, carpsucker, and buffalo were collected from the Clinch River and carp and buffalo were collected from the Tennessee River. The fish were pressure cooked and the flesh was separated from the bone for analysis.

Not all species of bottom feeders and sight feeders were collected in the sampling programs during 1960 to 1962. Therefore bottom feeders collected during 1960 to 1962 were considered as one sample; sight feeders were treated in similar manner. This evaluation of internal dose disregards any differences in fish due to the time of collection. Information on seasonal variation of such factors as feeding rates and water content of the flesh and their effect on radionuclide concentrations is unavailable and cannot be considered in the calculations.

Results of analyses of fish collected from the Clinch River and Tennessee River are listed in Tables 13 and 14, respectively. Average values are given for the principal radionuclides detected; analyses of the 1963 catch included only ^{90}Sr and ^{137}Cs. Variation of the averages is indicated by the standard error of the mean. Standard errors do not appear where fish were composited before analyses. Bottom feeders are listed by species, since information is available on the total quantities of these fish harvested commercially from Watts Bar Reservoir and from East Tennessee (Table 15). Information on sight feeders harvested is meager in comparison and does not warrant analyses by species. Only sport fishing takes place on the Clinch River.

Average values (for the same period) are observed to vary by factors ranging from about 2 to 5 between fish types from the same river; similar variations occur between fish of the same type but from the two rivers. A peculiar difference is noted in ^{90}Sr concentrations in sight feeders collected 1960-1962; the average concentration in Tennessee River fish is about 50% greater than in Clinch River fish. There is no explanation for this difference. Four carpsuckers, collected at Clinch River Mile 19.6,

TABLE 13

CONCENTRATION OF RADIONUCLIDES IN CLINCH RIVER FISH

(pc/kg fresh weight)

Fish Species	SAMPLE PERIOD	^{90}Sr Flesh	^{90}Sr Total[a]	^{137}Cs Flesh	^{137}Cs Total[a]	^{106}Ru Flesh	^{106}Ru Total[a]	^{60}Co Flesh	^{60}Co Total[a]
Carp	1960 - 1962	(17)[b] 500 ± 140	(40) 5100 ± 1700	(71) 510 ± 57	(39) 560 ± 79	(69) 170 ± 18	(39) 290 ± 78	(67) 66 ± 6.1	(39) 49 ± 9.9
	1963	(20) 91 ± 22		(20) 320 ± 110					
Carpsucker	1960 - 1962	(18) 540 ± 190	(39) 940 ± 120 (39) 4800[d]	(122) 1200 ± 460	(37) 640 ± 67	(22) 120 ± 30	(37) 56 ± 16	(22) 120 ± 19	(37) 32 ± 6.8
	1963	(20) 22 ± 4.4		(20) 460 ± 82					
Buffalo	1960 - 1962	(3) 240 ± 89	(30) 830 ± 110	(5) 480 ± 94	(30) 590 ± 92	(5) 110 ± 32	(30) 150 ± 38	(5) 78 ± 21	(30) 32 ± 6.8
	1963	(20) 43 ± 14		(21) 560 ± 84					
Sight Feeders[c]	1960 - 1962	(109) 180 ± 83		(126) 680 ± 120		(127) 120 ± 32		(127) 22 ± 11	

[a]Total fish consists of flesh and bone.

[b]Parenthetical values are numbers of fish analyzed.

[c]Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

[d]Includes four carpsuckers (composited) collected at CRM 19.6.

TABLE 14

CONCENTRATION OF RADIONUCLIDES IN FLESH OF
TENNESSEE RIVER FISH
(p c/kg Fresh Weight)

Fish Species	Sample Period	^{90}Sr	^{137}Cs	^{106}Ru	^{60}Co
Carp	1960-1962	(13)[a] 120 ± 33	(14) 180 ± 55	(14) 80 ± 27	(14) 71 ± 17
	1963	(20) 5.1 ± .75	(19) 61 ± 17		
Carpsucker	1960-1962	(10) 99 ± 28	(10) 130 ± 27	(10) 69 ± 23	(10) 62 ± 18
Buffalo	1963	(20) 8.9 ± 2.9	(20) 73 ± 12		
Sight Feeders[b]	1960-1962	(24) 250	(24) 170	(24) 48	(24) 66

[a]Parenthetical values are numbers of fish analyzed.

[b]Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

TABLE 15

COMMERCIAL FISH HARVEST FROM WATTS BAR RESERVOIR AND EAST TENNESSEE

(Pounds Fresh Weight)

Location	Carpsucker	Carp	Smallmouth Buffalo
Watts Bar Reservoir	15,600	23,700	161,000
East Tennessee	61,700	135,000	327,000
Fish Dilution Factor[a]	3.95	5.70	2.03

[a]Fish dilution factor = $\frac{\text{Pounds of East Tennessee Fish}}{\text{Pounds of Watts Bar Fish}}$

contained sufficient radioactivity to autoradiograph. This is typical of fish that have spent considerable time in White Oak Creek (or White Oak Lake).[30] Inclusion of these fish in the 1960-1962 analysis can be seen to increase significantly the average concentration of radionuclides. Although the concentrations of ^{90}Sr and ^{137}Cs in fish flesh during 1963 are found to be less than that observed during the period 1960 to 1962, a "t" test showed that only ^{90}Sr in carp and carpsucker and ^{137}Cs in carp are significantly different at the 5% level. These smaller concentrations of ^{90}Sr and ^{137}Cs in fish flesh are attributed to the reduction of radionuclide release to the Clinch River. Not to be overlooked is the fact that fallout from nuclear tests contributes about 45% of ^{90}Sr and 20% of ^{137}Cs to the total load in the Clinch River during 1962 and 1963.

Estimated Intake of Radionuclides from Fish

An estimate is made of man's intake of radionuclides (including radioactive fallout) by assuming an annual rate of fish consumption of 37 lb.[31] This rate of fish consumption applies to commercial fishermen and, as a result, the estimate of intake is for an admittedly high exposure group. Data on the quantity of specific types of fish consumed are not available. A recent survey of 80 fishermen by the Tennessee Fish and Game Commission (completed August 1964) reports that fish are consumed with one meal each week. None of those interviewed prepare the total fish (flesh plus bone) for eating.

Calculations made are based on an annual consumption of 37 lb of bottom feeders, considering both the total fish and the flesh, and consumption of 37 lb of sight feeders, considering only the flesh. The fraction of the various species of bottom feeders caught is assumed to be distributed according to commercial harvests from Watts Bar Reservoir. Estimates of the annual intake of specific radionuclides by consuming Clinch River or Tennessee River fish are given in Table 16 and 17, respectively. A very noticeable increase in ^{90}Sr intake is observed when consumption of bottom feeders (total fish) is considered. This significantly larger intake is due to the concentration of ^{90}Sr by the bones of the fish, all of which are assumed to be eaten. The assumption that 37 lb of total fish are consumed each year is certainly conservative. However, without better data on fish consumption, it is impossible to arrive at a more reasonable value of intake.

Radionuclide intake by the general population is likely to be influenced from dilution by other fish harvested in East Tennessee, as well as by differences in radionuclide content among species of bottom feeders. Applying a fish dilution factor (bottom feeders) for East Tennessee fish (Table 15), gives the revised annual intakes shown in Tables 16 and 17. A significant decrease in radionculide intake is observed, the reduction factors ranging from about 2 to 4. Fish collected from the Clinch River or the Tennessee River and shipped outside the East Tennessee area are likely to be diluted even more with fish from other parts of the country.

Relation to Permissible Intake for Man.- A maximum permissible intake (MPI) is estimated by assuming a daily intake of 2.2 liters of water containing the maximum permissible concentration (MPC) of the radionuclide of interest. Using the estimated intakes (Tables 16 and 17), it is possible to calculate the fraction of MPI attained as a result of eating contaminated fish (Tables 18 and 19). The estimates indicate that bone of the highest exposure group receive the largest dose; on the average, 7.0 to 8.6% of the MPI is attained as a result of consuming bottom feeders (total fish) from the Clinch River. Strontium-90 is responsible essentially for the total bone dose. If only the flesh of bottom feeders is consumed, the percentage of MPI attained is reduced to 1.5%. As shown, further reduction in dose is likely due to dilution with other East Tennessee fish. The estimated percentage of MPI attained during 1963 is less than 1% for the critical organs, bone, total body, GI tract, and thyroid.

More information would be required to estimate the intake of radionuclides due to past events. Such information would include: (1) rate of transfer of radionuclides from water to fish (flesh and bone) as a function of radionculide and stable element concentration, fish age, and season of the year: (2) rate of transfer of radionuclide from bone to flesh while cooking and method of fish preparation; and (3) type and quantity of fish consumed and fish eating habits of individuals as a

TABLE 16

ESTIMATED ANNUAL INTAKE OF RADIONUCLIDES BY ASSUMED CONSUMPTION OF CLINCH RIVER FISH[a]

(nc/year)

Fish	Sample	^{90}Sr		^{137}Cs		^{106}Ru		^{60}Co	
Species	Period	Flesh	Total[b]	Flesh	Total[b]	Flesh	Total[b]	Flesh	Total[b]
Bottom Feeders[c]	1960-1962	4.9±1.3	23±3.7 (28)[f]	9.0±1.4	10±1.3	2.0±0.44	2.7±0.54	1.3±0.28	0.58±0.94
Bottom Feeders[c]	1963	0.78±0.19		8.8±1.2					
Bottom Feeders[d]	1960-1962	1.9±0.60	7.6±0.83 (8.9)	3.7±0.64	4.4±0.19	0.81±0.21	1.1±0.25	0.58±0.44	0.24±0.061
Bottom Feeders[d]	1963	0.32±0.091		3.9±0.56					
Sight Feeders[e]	1960-1962	3.0±1.4		11±2.6		2.0±0.54		0.38±0.19	

[a]Calculated from the concentration of the radionuclide found in either bottom feeder flesh, bottom feeder total fish, or sight feeder flesh, and an assumed consumption of 37 lbs per year of each category. Thus, these calculated intakes are not additive.

[b]Total fish consists of flesh and bone.

[c]Bottom feeders include carp, carpsucker, and buffalo.

[d]Intake adjusted by fish dilution factor.

[e]Sight feeders include white crappie, blue gill, white bass, largemouth bass, sauger, and drum; catfish also included.

[f]Parenthetical values include four carpsuckers (composited) collected at CRM 19.6.

TABLE 17

ESTIMATED ANNUAL INTAKE OF RADIONUCLIDES BY ASSUMED CONSUMPTION OF FLESH OF TENNESSEE RIVER FISH[a]
(nc/year)

Fish Species	Sample Period	^{90}Sr	^{137}Cs	^{106}Ru	^{60}Co
Bottom Feeders[b]	1960-1962	1.9 ∓ 0.38	2.7 ∓ 0.59	1.3 ∓ 0.31	1.1 ∓ 0.21
Bottom Feeders[c]	1963	0.14 ∓ 0.042	1.2 ∓ 0.18		
Bottom Feeders[d]	1960-1962	0.39 ∓ 0.075	0.53 ∓ 0.11	0.26 ∓ 0.062	0.23 ∓ 0.043
Bottom Feeders[d]	1963	0.066 ∓ 0.021	0.55 ∓ 0.085		
Sight Feeders[e]	1960-1962	4.3	2.8	0.81	1.1

[a]Calculated from the concentration of the radionuclide found in either bottom feeder flesh or sight feeders flesh and an assumed consumption of 37 lbs per year of each category. Thus, these calculated intakes are not additive.

[b]Bottom Feeders include carp and carpsucker.

[c]Bottom Feeders include carp and buffalo.

[d]Intake adjusted by Fish Dilution Factor.

[e]Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

TABLE 18

ESTIMATED PERCENTAGE OF MPI THAT MAN MAY ATTAIN BY CONSUMING CLINCH RIVER FISH[a]

Fish Species	Sample Time	Critical Organ			
		Bone	Total Body	GITract	Thyroid
Bottom Feeders[b] (flesh)	1960-1962	1.5∓0.39	0.87∓0.23	0.072∓0.0081	0.38∓0.072
Bottom Feeders[b] (flesh)	1963	0.27∓0.059	0.19∓0.034	0.030∓0.0035	0.060∓0.010
Bottom Feeders[b] (total[c])	1960-1962	7.0∓1.1 (8.6)[f]	4.1∓0.66 (5.0)	0.14∓0.014 (0.15)	1.4∓0.19 (1.6)
Bottom Feeders[d] (flesh)	1960-1962	0.60∓0.19	0.36∓0.11	0.03∓0.0039	0.16∓0.034
Bottom Feeders (flesh)[d]	1963	0.11∓0.028	0.081∓0.016	0.013∓0.0018	0.024∓0.0049
Bottom Feeders[d] (total)	1960-1962	2.4∓0.28 (2.9)[f]	1.4∓0.19 (1.7)	0.053∓0.0047 (0.0058)	0.48∓0.051 (0.57)
Sight Feeders[e] (flesh)	1960-1962	0.94∓0.43	0.61∓0.25	0.071∓0.012	0.31∓0.080

[a]The ratio of the estimated annual intake of radionuclides from consuming the particular category of fish to the maximum permissible intake (MPI) for the critical organ of interest. Thus these calculated percentages of MPI are not additive.

[b]Bottom feeders include carp, carpsucker, and buffalo.

[c]Total fish consist of flesh and bone.

[d]Intake adjusted by Fish Dilution Factor.

[e]Sight Feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

[f]Parenthetical values include four carpsuckers (composited) collected at CRM 19.6.

TABLE 19

ESTIMATED PERCENTAGE OF MPI THAT MAN MAY ATTAIN BY CONSUMING FLESH OF TENNESSEE RIVER FISH[a]

Fish Species	Sample Period	Critical Organ			
		Bone	Total Body	GI Tract	Thyroid
Bottom Feeders[b]	1960-1962	1.8 ∓ 0.36	3.7 ∓ 0.68	0.11 ∓ 0.014	0.55 ∓ 0.084
Bottom Feeders[c]	1963	0.14 ∓ 0.039	0.33 ∓ 0.075	0.012 ∓ 0.0020	0.029 ∓ 0.0066
Bottom Feeders[d]	1960-1962	0.37 ∓ 0.071	0.69 ∓ 0.14	0.021 ∓ 0.0026	0.11 ∓ 0.017
Bottom Feeders[d]	1963	0.066 ∓ 0.019	0.15 ∓ 0.037	0.0057 ∓ 0.00082	0.013 ∓ 0.0035
Sight Feeders[e]	1960-1962	4.0	7.6	0.11	0.83

[a]The ratio of the estimated annual intake of radionuclides from consuming the particular category of fish to the maximum permissible intake (MPI) for the critical organ of interest. Thus, these calculated percentages of MPI are not additive.

[b]Bottom feeders include carp and carpsucker.

[c]Bottom feeders including carp and buffalo.

[d]Intake adjusted by Fish Dilution Factor

[e]Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, and drum; catfish also included.

function of age. Current research suggests that the concentration of ^{90}Sr in the flesh of white crappie rapidly reaches equilibrium with the water.[32] Such information helps to answer, in part, the questions raised by (1) above and an extension of such studies will enhance the estimates of dose to man by this path of intake.

An interesting calculation was made based on the ^{90}Sr content of the four carpsuckers previously mentioned. The combined weight of flesh and of whole fish (flesh and bone) for the four carpsuckers was 0.51 kg and 0.85 kg, respectively; the ^{90}Sr concentration in flesh was 500 pc/kg and in whole fish was 43000 pc/kg. An individual eating the four fish could have attained 0.1% of MPI (bone) from the flesh and 11% of MPI from the whole fish. Although such an event is unlikely, it indicates the possibility that consumption of relatively few fish (flesh and bone) could lead to a significant exposure.

Computer Calculations of Internal Dose

The dose received by the skeleton, total body, and thyroid of man, due to consumption of contaminated water and fish, is calculated by use of the models described in the section, "Estimation of Radiation Dose Following Ingestion of Contaminated Water". In addition to the assumptions listed for contaminated drinking water, it is also assumed that 37 lb per year of the flesh of bottom feeders is consumed by a standard man during the period 1960 to 1963. Without information on actual fish consumption as a function of age, it is further assumed that the intake of fish is distributed as the intake of water. Another way to state this assumption is that the ratio of fish eaten by an individual to that of standard man is equal to the ratio of water consumed by the individual to that of standard man.

Figure 9 shows the computed annual dose to the skeleton due to consumption of contaminated water and fish. By comparison with Figure 6 it is seen that the net increase in dose rate to the skeleton is small. This is due to the fact that data for only four years of fish collection (1960-1963) is available for the calculations, to the long effective half life of the critical radionuclide, ^{90}Sr and to the reduction in ^{90}Sr released to the river. The net increase in total dose received through

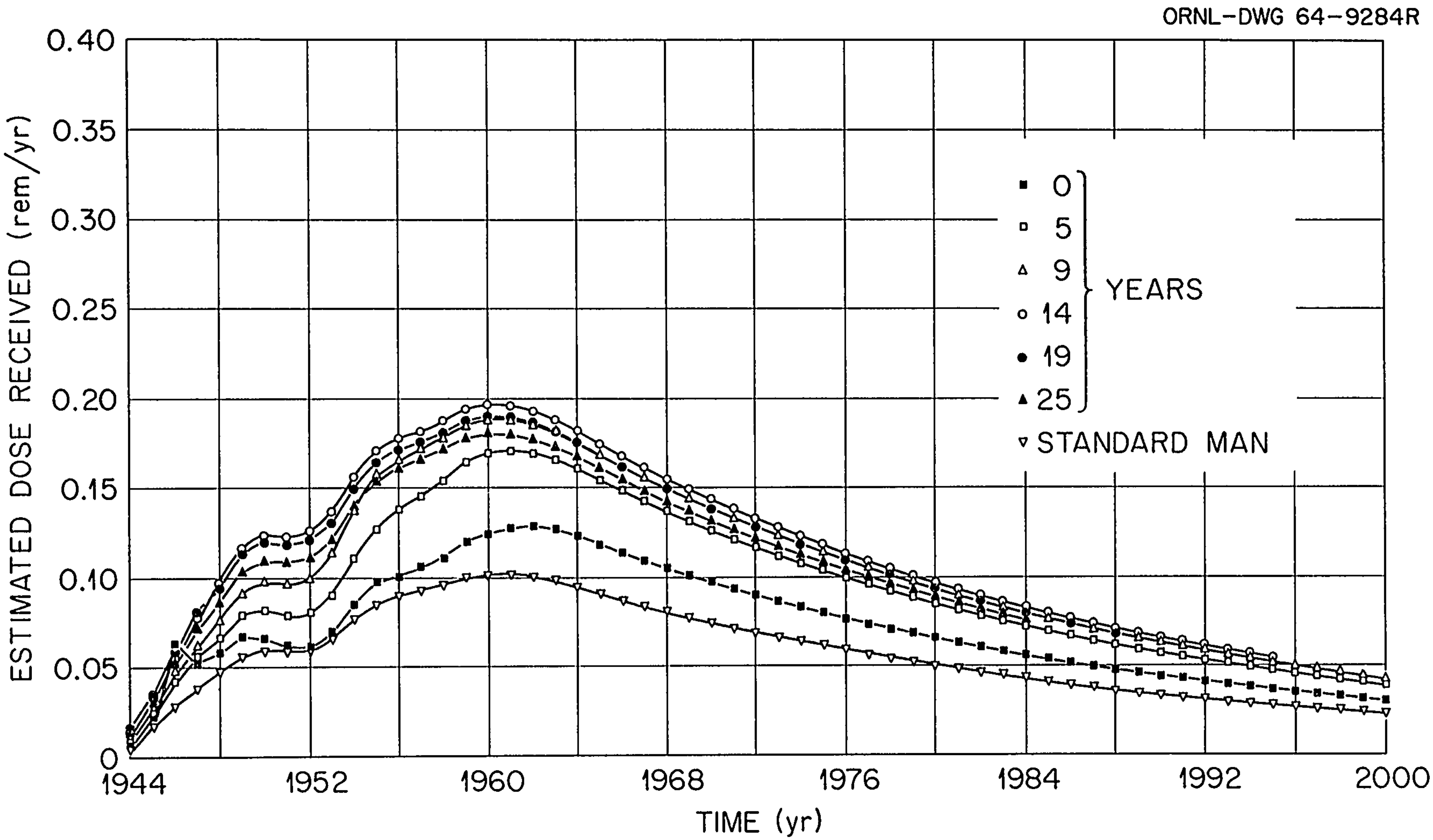


Fig. 9. Estimated Dose Received by Skeleton of Males from Drinking Clinch River Water and Consuming Contaminated Fish.

1963 by the organs of interest is given in Table 20. The cumulative dose over the 4-year exposure period is not excessive, with the skeleton receiving the largest increase of about 30 mrad. Consumption of total fish could result in an increase of the cumulative dose by a factor of 5 to 10, but available information does not justify such an assumption.

Exposures from External Sources

Radionuclides in Water Treatment Plants

The presence of radionuclides in raw water entering a water treatment plant may lead to their concentration in the plant and create an external or internal dose problem. Three water systems using Clinch River water as a source of supply were investigated. The Oak Ridge Water Plant has its raw water intake at CRM 41.5, well above the outfall of White Oak Creek. The other two water treatment plants - serving the Oak Ridge Gaseous Diffusion Plant (ORGDP) and the Kingston Steam Plant - have water intakes at CRM 14.5 and on the Emory River near CRM 4.4, respectively. These water treatment plants are basically similar in design. The treatment processes include: prechlorination for algae control; coagulation using alum, soda ash (as dictated by raw water alkalinity), and occasionally coagulant aids for turbidity removal; settling; filtration (either sand or anthracite media); and postchlorination for disinfection. Activated carbon is used when taste and odor problems occur. Water used in boilers is treated further by zeolite softeners.

The investigation consisted of: external radiation surveys, using a scintillation-type survey meter (calibrated with radium); collection and analysis of samples of sludge from settling basins, condensers, hot water heaters, boilers, air conditioners, and an elevated tank; collection and analysis of samples of sediment from filters and cores of filter media; and collection and analysis of samples of zeolite softener regenerant, as well as the softener media.

At the time of the surveys, various amounts of water had been treated since the last time settling basins had been cleaned or filters backwashed (Table 21). Thus, there was variation in the amount of accumulated sludge in the settling basins and sediment on the filters. Results of the external radiation survey are summarized in Table 22. Generally there was little

TABLE 20

DOSE RECEIVED BY CRITICAL ORGANS OF MALES FROM CONSUMING FISH[a]
(rem)

Age at Start of Exposure - 1944	Clinch River Water			Tennessee River Water		
	Skeleton	Total Body	Thyroid	Skeleton	Total Body	Thyroid
0	0.033	0.0035	0.011	0.013	0.0014	0.0037
5	0.031	0.0032	0.0089	0.012	0.0012	0.0031
9	0.028	0.0029	0.0078	0.011	0.0011	0.0027
14	0.030	0.0026	0.0072	0.011	0.0011	0.0025
19	0.027	0.0026	0.0069	0.011	0.0010	0.0024
25	0.026	0.0025	0.0066	0.010	0.0010	0.0023
Standard Man	0.015	0.0021	0.0047	0.006	0.0008	0.0016

[a]The cumulative dose for the period 1960-1963.

TABLE 21

OPERATIONAL DATA OF WATER TREATMENT PLANTS

System	Volume Through[a] Flocculator and Settling Basin (gal)	Volume Through[a] Filter (gal)	Sludge in Settling Basin (cu ft)	Plant Capacity (gal/day)
Oak Ridge Water Plant	1.1×10^9	1.8×10^6	2×10^4	22×10^6
ORGDP	5.4×10^8	4.5×10^6	6×10^3	4×10^6
Kingston Steam Plant	1.9×10^6	3.7×10^5		5.7×10^5

[a]Volume through flocculator, settling basin, and filter since last cleaned.

TABLE 22

MEASUREMENTS OF IONIZING RADIATION IN WATER TREATMENT PLANTS

(mr/hr)

System	Ground Surface	Flocculator	Settling Basin	6 in. Above Water in Settling Basin	Filter
Oak Ridge Water Plant	0.016	0.013	0.012	0.0097	0.0095
ORGDP	0.017	0.011	0.012	0.0092	0.0092
Kingston Steam Plant	0.015	0.0083	0.0087		0.015

[a] All measurements (except as noted) were made 3 ft above walking surface of the particular component of the treatment plant.

difference in the dose rates at different units of the plants. Dose rates above background levels (the Oak Ridge Water Treatment Plant was considered as background) were, with one exception, not found. At a distance of 2 in. above a partially drained filter at the Kingston Steam Plant, a dose rate of 0.021 mr/hr was measured. The dose rate remained the same after the filter was backwashed. It is likely that the anthracite media of the filter to some extent concentrates radionuclides by sorption. The dose rate above these filters (0.015 mr/hr) was also influenced by the natural radioactivity present in block used for construction of the building. External exposure to radioactive materials concentrated in these water treatment plants was not significantly different from exposure to background radiation.

Immersion in Contaminated Water

Due to the presence of radionuclides, the river will act as a source of radiation to persons engaged in swimming, boating, fishing, and water skiing. Since direct measurements of immersion dose rate were unavailable, it was necessary to estimate the dose rate by considering the radionuclide composition of the water.

The immersion dose calculation assumes the body is in the center of a sphere and receives equal quantities of radiation from all directions. The external exposure from beta radiation may be written in units of rad per day:[33]

$$\text{Beta Dose Rate} = \frac{3.7 \times 10^{4}\ \frac{\text{dis/sec}}{\mu\text{c}} \times 8.64 \times 10^{4}\ \frac{\text{sec}}{\text{day}} \times Q\ \frac{\mu\text{c}}{\text{g}} \times a \times b \times E_m \times 10^{6}\ \frac{\text{ev}}{\text{Mev}} \times P_t}{6.25 \times 10^{13}\ \frac{\text{ev}}{\text{g-rad}} \times P_m \times N} \qquad (15)$$

where

$Q = \mu c/g$ of water

a = factor introduced in case the radius of beta emitting medium is less than the maximum range of the beta ray,

$b = \frac{E_{avg}}{E_m}$,

P_t = relative mass stopping powerof tissue,

P_m = relative mass stopping power of water,

$N = \frac{\text{ev/ion pair}}{32.5}$,

E_m = maximum energy of type considered, and

E_{avg} = average energy of type considered.

Assuming that $a = 1$, $N = 1$, $P_t = P_m$, and $E_{avg} = E_i$ (effective absorbed energy per disintegration), the expression is simplified to:

$$\text{Beta Dose Rate} = 51.2\ QE_i \qquad (16)$$

An empirical formula was used to estimate the average effective absorbed energy of a beta disintegration.[1]

$$E_i = 0.33\ E_m\ f \left(1 - \frac{\sqrt{z}}{50}\right) \left(1 + \frac{\sqrt{E_m}}{4}\right) \qquad (17)$$

where

f = fraction of disintegration at a particular energy,

z = atomic number

The penetration distance in water of the most energetic beta particles from the radionuclides involved is about one centimeter. Therefore, the beta radiation at the surface of a body immersed in the contaminated water is effectively one-half of that calculated by equation 16.

Similarly, the external exposure from gamma radiation may be written in units of rad per day:[33]

$$\text{Gamma Dose Rate} = \frac{51.2\ Q\ E\ Pt\ (\mu - \sigma s)_m (1-e^{-\mu_\ell r})}{P_m\ \mu_\ell} \qquad (18)$$

where

μ = total absorption coefficient,

σ_s = Compton scattering coefficient,

μ_ℓ = $(\mu - \sigma s)_m$ plus the fraction of σ_s representing the scattered and degraded radiation that reaches the point of measurement,

r = radius of contaminated medium,

$E = E_m\ f(1-e^{-\sigma x})$ (from ICRP 1959),

σ = total absorption coefficient less Compton scattering coefficient in body organ for the given photon energy,

x = effective radius of body organ.

Assuming that $P_t = P_m, \mu_\ell = (\mu - \sigma s)_m$, $r = \infty$, σx is large, and that the submerged body is receiving radiation from 4 π steradians, the expression becomes:

$$\text{Gamma Dose Rate} = 51.2\ QE_m f \qquad (19)$$

In each instance where some latitude is allowed in the assumptions, a conservative approach was taken. Therefore the computed dose rates are judged to be conservative.

Where the water contains a mixture of radionuclides, it is necessary to calculate the dose rate from each radionuclide. The total dose rate is then the sum of the individual dose rates. Decay schemes presented by Blomeke and Todd were used in the calculations.[34] To simplify calculations, the dose rate of each radionuclide was normalized for a concentration of one μ c per ml (Table 23). Tabulated values are one-half the beta dose rate (equation 16) and the total gamma dose rate (equation 19).

The immersion dose rates due to beta and gamma radiation at the two stations are listed in Table 24 and shown graphically in Fig. 10 for CRM 14.5. The total dose rate at CRM 14.5 and TRM 465.5 is shown in Fig. 11.

TABLE 23

IMMERSION DOSE RATE OF RADIONUCLIDES

(rad/day per μc/ml)

Nuclide	Beta	Gamma	Total	Parent Plus Daughter
Co^{60}	2.90	128	131	131
Sr^{90}	5.4	0	5.4	
Y^{90}	22.1	0.29	22.4	27.8
Cs^{137}	4.97	0	4.97	
Ba^{137}	0	33.8	33.8	38.8
Ru^{106}	0.30	0	0.30	
Rh^{106}	33.6	12.6	46.2	46.5
Zr^{95}	3.25	36.0	39.3	39.3
Nb^{95}	0.13	38.1	38.2	38.2
Ce^{144}	2.11	2.06	4.17	
Pr^{144}	29.6	4.06	33.7	37.9
Y^{91}	14.9	0.19	15.1	15.1
I^{131}	4.93	19.7	24.6	24.6

TABLE 24

IMMERSION DOSE RATES IN CLINCH AND TENNESSEE RIVERS

(units of 10^{-4} mrad/24-hr Exposure)

Year	Clinch River Mi 14.5			Tennessee River Mi 465.5		
	Beta	Gamma	Total	Beta	Gamma	Total
1949	19	16	35	2.4	2.0	4.4
1950	3.4	5.2	8.6	0.5	0.79	1.3
1951	2.7	2.1	4.8	0.46	0.35	0.81
1952	8	5.0	13	1.3	0.77	2.1
1953	13	2.7	16	2.0	0.41	2.4
1954	20	7.2	27	2.3	0.82	3.1
1955	18	9.9	28	2.8	1.6	4.4
1956	16	18	34	2.6	2.9	5.5
1957	10	9.9	20	1.4	1.4	2.8
1958	16	8.4	24	3.1	1.6	4.7
1959	71	67	140	8.5	8.0	17
1960	170	95	270	25	14	39
1961	160	79	240	21	10	31
1962	89	38	120	12	5.2	17
1963	33	17	50	4.6	2.3	6.9

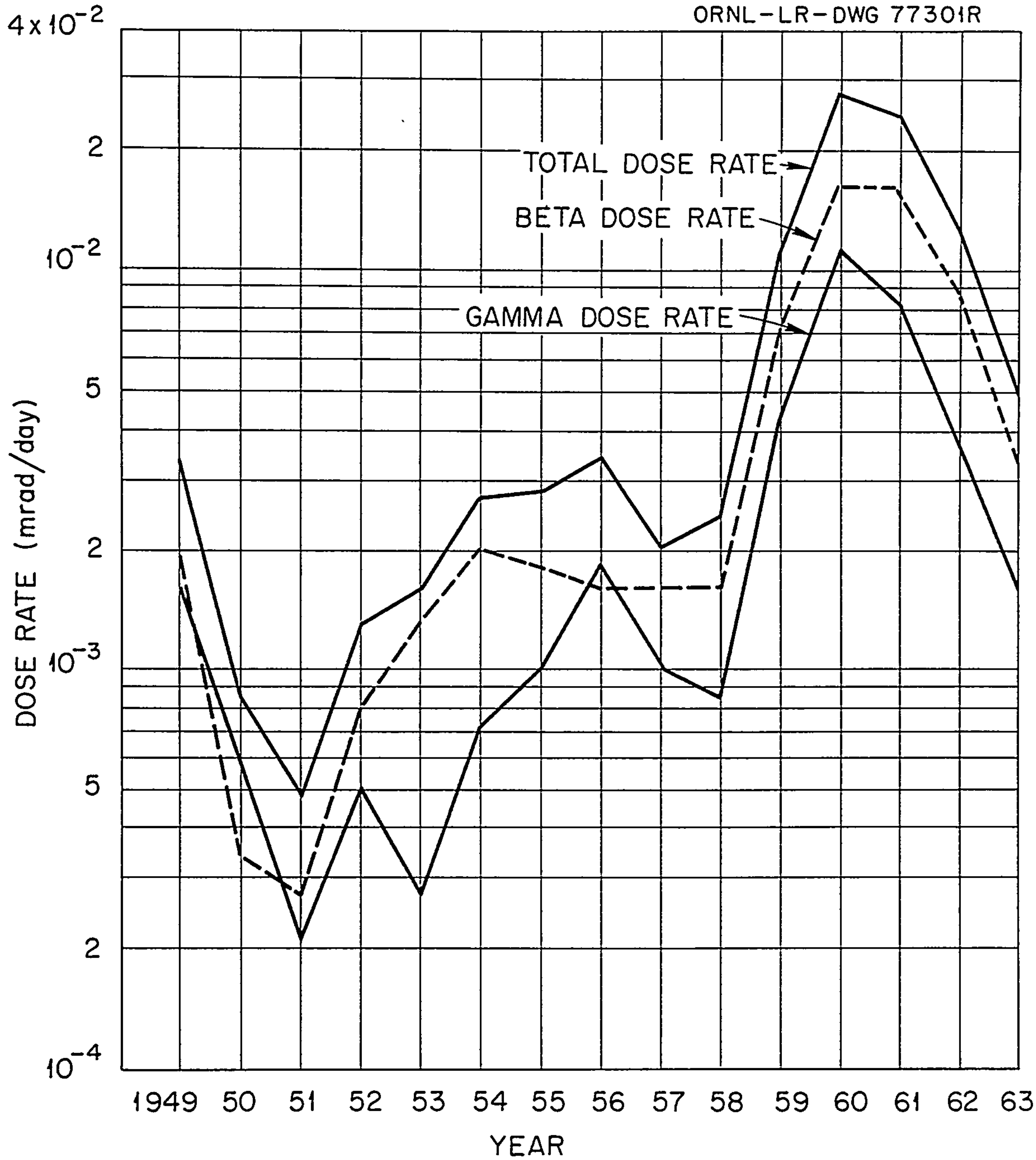


Figure 10. Immersion Dose Rate at CRM 14.5

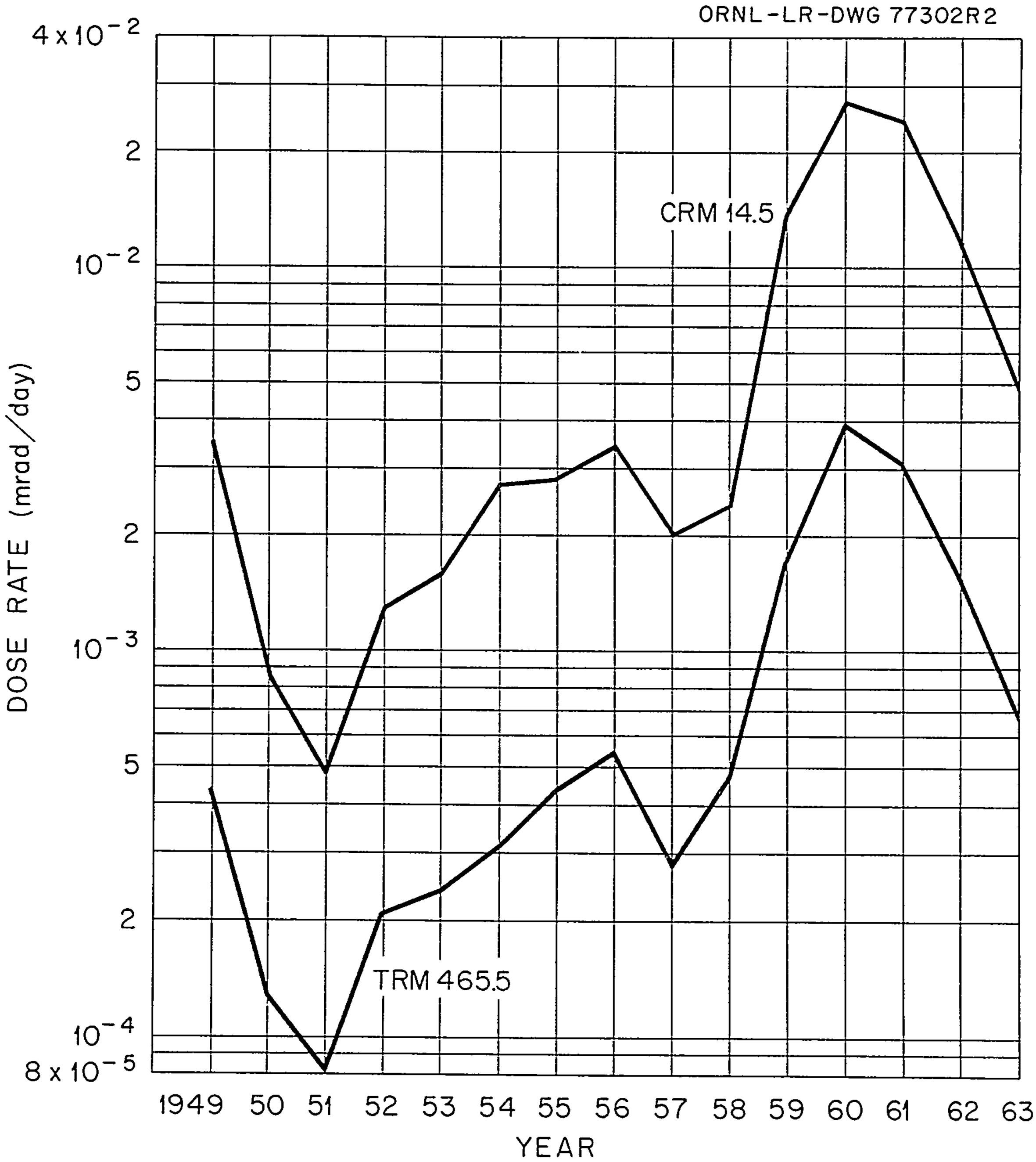


Fig. 11. Total Immersion Dose Rate at CRM 14.5 and TRM 465.5

A maximum dose rate of 0.027 mrad per day of exposure at CRM 14.5 (1960) is estimated. The dose rate is a function of nuclide type and concentration. Until 1958, the largest fraction of beta dose was associated with ^{90}Sr and the largest gamma dose was generally due to ^{137}Cs. Since then, ^{106}Ru has accounted for about 75% of the total immersion dose.

Contaminated Bottom Sediments

Radionuclides associated with solids that have settled to the bottom of the river can be expected to contribute to the total dose received by man. Although earlier calculations assumed complete dilution of fission products in the river, annual surveys made by the ORNL Applied Health Physics Section show that some of the radionuclides are retained by the bottom sediments.[35]

Radionuclide Concentrations in Sediments.-- Measurements were made at cross sections 2 miles apart in the Clinch River and at 50-ft intervals across the river at each cross section. In the Tennessee River and TVA Reservoirs, measurements were made at sections approximately 10 miles apart and at 10 points at each station. Measurements consisted of gamma counts obtained with a multiple-GM-tube detector ("Flounder"), lowered to the surface of the bottom sediments, and analysis of the mud samples taken at each measurement point. Average concentrations of specific radionuclides in bottom sediments were calculated by averaging all values for the entire reach of the lower Clinch River and of the Tennessee River (Table 25). Cesium-137, ^{144}Ce, ^{60}Co, and ^{106}Ru, were found to be the principal radionuclides associated with these sediments. Reasons for such selectivity are enumerated elsewhere.[36] The values listed as ^{91}Y are not measured but, as mentioned earlier, are calculated as the difference between concentrations of trivalent rare earths and ^{90}Sr. The reasons for large changes in concentrations of ^{137}Cs and ^{106}Ru in sediments have already been discussed.

The "Flounder" is used principally to furnish qualitative information on the build-up of gamma emitting radionuclides in bottom sediments. Construction of the device makes it insensitive to beta radiation. Although the "Flounder" is calibrated routinely with a sealed radium source, the complex spectrum of gamma rays from both the contaminated sediments

TABLE 25

AVERAGE CONCENTRATION OF RADIONUCLIDES IN BOTTOM SEDIMENTS[a]

Nuclide	1954	1955	1956	1957	1958	1959	1960	1961
				CLINCH RIVER				
Cs^{137}	20	25	200	210	160	280	170	81
Sr^{90}	3.6	3.8	6.0	4.1	5.9	5.4	2.4	0.85
Ce^{144}	6.1	24	41	12	22	38	19	6.9
Y^{91}	0.8	5.9	7.8	1.5	6.8	83	79	20
Ru^{106}	4.5	4.8	8.1	5.6	8.6	12	70	130
Co^{60}	19	21	42	15	12	33	19	11
				TENNESSEE RIVER				
Cs^{137}	6.6	6.7	35	32	21	17	18	14
Sr^{90}	2.4	0.3	2.5	0.76	1.4	0.8	0.5	0.41
Ce^{144}	1.6	11	8.3	2.7	6.6	4.5	1.6	1.5
Y^{91}	0	5.9	1.2	0.84	4.3	4.3	5.6	2.8
Ru^{106}	1.5	2.7	3.0	2.3	3.5	4.6	15	23
Co^{60}	5.8	8.0	7.0	3.6	3.3	3.7	3.2	2.4

[a] 10^{-6} μ c/gram DRIED SEDIMENT.

and the radium source prevents a direct determination of exposure dose by use of this instrument. Estimates of exposure dose can be made (Tables 26 and 27) but it is necessary to recognize their limitations. Figure 12 shows the average gamma counting rate in the Clinch River and Tennessee River, as determined by the "Flounder" and averaged for the entire study reach of each, and the curies per year of ^{137}Cs and ^{60}Co released. In general, measurements in the Clinch River reflect the quantity of ^{137}Cs and ^{60}Co released each year. Maximum readings in the Clinch River (generally at CRM 8.3) were larger than the average readings by a factor of 1.9 ± 0.09; similarly, this ratio in the Tennessee River was 1.8 ± 0.2.

Estimation of Radiation Dose from Sediments. - For the purpose of estimating the radiation dose, it was assumed that the average radionuclide composition of the sediments was uniformly distributed in an infinite source. Further, it was assumed that the individual would be exposed to one-half the submersion dose of beta particles and gamma photons (i.e., from one-half a sphere). Such an assumption is reasonable, since the exposed individual is likely to be standing on or floating above the contaminated sediments. Normally, only the feet would be subjected to the total beta dose rate and some fraction greater than one-half of the gamma dose rate.

Estimated dose rates from bottom sediments in the Clinch River and Tennessee River are listed in Tables 26 and 27. The beta dose rate was taken as one-half the value determined by use of equation 16 and the gamma dose rate by use of one-half the calculated value of equation 19. Since the source is not infinite in extent, the calculated values of gamma dose rate are overestimates. Accordingly, the highest dose rate of 12 mrad per day occurred in 1959, and was divided as 40% beta and 60% gamma radiation. The percentage contributions of specific radionuclides to the beta and gamma dose rates are listed in Table 28. The total rare earths, ^{137}Cs, and ^{106}Ru are the principal contributors to beta dose rates, and ^{60}Co and ^{137}Cs account for the largest fraction of gamma dose rate.

TABLE 26

ESTIMATED RADIATION DOSE RATES FROM CONTAMINATED SEDIMENTS IN CLINCH RIVER

	Measured[a] (10^{-2} mr/24-hr)		Calculated (10^{-2} mrad/24-hr exposure)			
Year	Average	Maximum	Beta	1/2 Gamma[c]	Total	Attenuated[b] 1/2 Gamma[c]
1951	39			90[d]		
1952	88			320[d]		
1953	53			160[d]		
1954	57	110	60	160	220	9.5
1955	60	110	130	180	310	11
1956	130	260	300	630	930	35
1957	96	180	180	460	640	24
1958	100	200	210	360	570	19
1959	160	280	450	710	1160	39
1960	150	280	510	460	970	25
1961	95	170	530	290	820	15

[a]In units of 10^{-2} mr/24-hr exposure as measured by the "Flounder."

[b]Attenuation through 3 ft of water.

[c]One-half of total gamma dose from infinite source.

[d]Estimated from correlation relationship.

TABLE 27

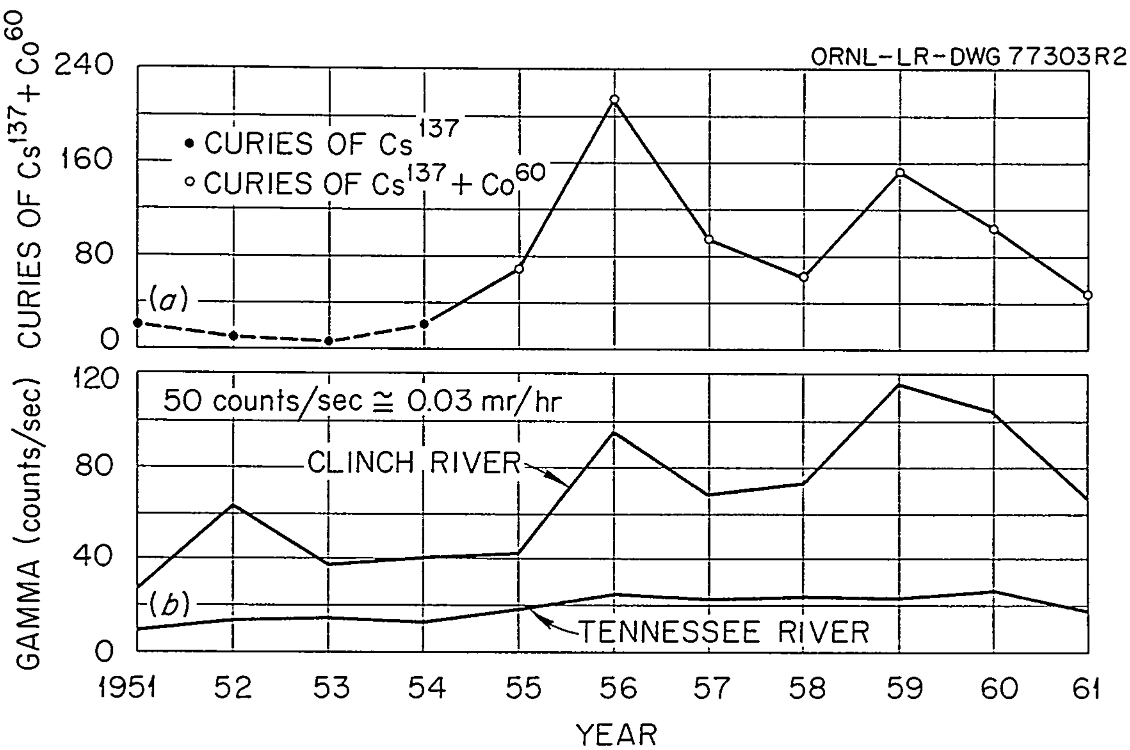
ESTIMATED RADIATION DOSE RATES FROM CONTAMINATED SEDIMENTS IN TENNESSEE RIVER

	Measured[a] (10^{-2} mr/24-hr)		Calculated (10^{-2} mrad/24-hr exposure)			
Year	Average	Maximum	Beta	1/2 Gamma[c]	Total	Attenuated[b] 1/2 Gamma[c]
1951	13					
1952	22					
1953	23					
1954	19	30	22	50	72	3.0
1955	26	43	60	68	128	4.2
1956	36	69	65	110	175	6.1
1957	33	58	37	80	117	4.2
1958	35	63	55	62	117	3.5
1959	30	63	48	56	104	3.1
1960	33	49	75	61	136	3.3
1961	26	48	95	54	149	2.8

[a]In units of 10^{-2} mr/24-hr exposure as measured by the "Flounder."

[b]Attenuation through 3 ft of water.

[c]One-half of total gamma dose from infinite source.



(a) CURIES OF Cs^{137} AND Co^{60} RELEASED TO CLINCH RIVER.

(b) AVERAGE GAMMA COUNT AT SURFACE OF BOTTOM SEDIMENTS.

Fig. 12. Comparison of Gamma Radiation from Bottom Sediments with Release of Cs^{137} and Co^{60} in Waste.

TABLE 28

PERCENTAGE CONTRIBUTION TO BOTTOM SEDIMENT DOSE RATE BY RADIONUCLIDES

Type of Radiation	Nuclide	CLINCH RIVER		TENNESSEE RIVER	
		1954-1959	1960-1961	1954-1959	1960-1961
Beta	TRE[a]	50	23	52	15
	Ru^{106}	14	64	22	75
	Cs^{137}	31	13	21	10
Gamma	Co^{60}	44	25	50	31
	Cs^{137}	53	55	45	47
	Ru^{106}	1	14	3	22

[a]TOTAL RARE EARTHS

An estimate can be made of the bottom sediment gamma dose rate in the Clinch River for periods when only "Flounder" measurements were made. This made possible by the apparent relationship between "Flounder" measurements and calculated gamma dose rates, and is expressed as a coefficient of correlation of 0.90. With "Flounder" measurements X as abcissa and gamma dose rates Y as ordinates, the relationship is given by the equation $Y = -0.84 + 4.64\ X$. The 95% confidence limits of the slope of the regression curve are ± 2.31. The correlation coefficient for similar data from the Tennessee River is 0.58, and the slope of the regression curve and its 95% confidence limits is 0.19 ± 3.45. Thus, estimates of bottom sediment gamma dose rates by use of the "Flounder" on the Tennessee River are not justified with the data available.

Since bottom sediments are generally covered by water, the gamma dose rate to the gonads of an individual standing on the river bottom would be reduced by attenuation. An average attenuation coefficient for water was calculated by weighting both the fraction of a disintegration that leads to a photon of a given energy from a particular radionuclide and the fraction each radionuclide contributes to the total loading of the bottom sediments. The fraction of dose remaining is graphed as a function of the depth of water (Fig. 13). The estimated gamma dose rates after attenuation through 3 feet of water are listed in Tables 26 and 27.

Cumulative Aggregate Dose to Exposed Populations

The aggregate exposure dose of individuals or critical population groups that resulting from disposal of radioactive waste to the Clinch River can not be estimated precisely. The principal reasons for this is the lack of information on habits and characteristics of the potentially exposed groups. Data on location and age distribution of potentially exposed populations, amounts of important foodstuffs consumed, methods of food preparation, and principal recreational habits are needed to define the total exposure dose. Age differences in metabolic rates or processes of children or adults as they relate to the important radionuclides, differences in radionuclide removal from river water by suspended solids and by water treatment processes, and differences in the transfer of

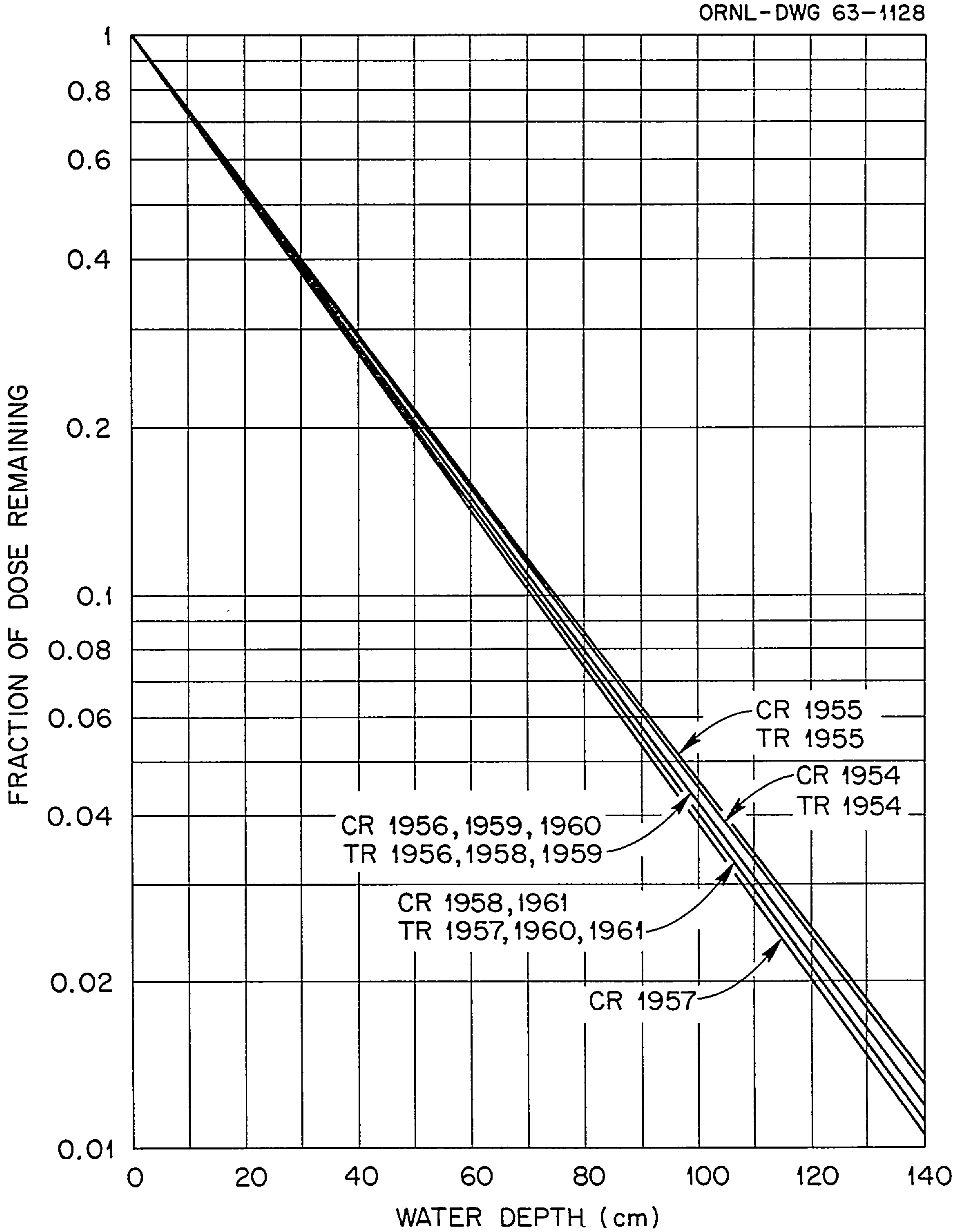


Figure 13. Attenuation of Bottom Sediment Dose in Clinch River and Tennessee River.

radionuclides from contaminated water to fish must also be considered. Although a single critical population group may be defined for a particular exposure pathway there is no reason to postulate the same critical population group for all exposure pathways.

By selecting reasonable values for periods of occupancy and dietary habits, an estimate can be made of the aggregate exposure dose (from 1944 to 1963) to the skeleton and total body of males working and residing in the Clinch-Tennessee River environment (Table 29). The fractions of allowable dose received by the thyroid and GI tract is smaller than that received by the total body and is not included. Since the Clinch River does not serve as a source of municipal water, children do not consume this water directly. Therefore, it is assumed that the youngest age group at the beginning of exposure is the 18-year old employed at the ORGDP. Only one-half of the daily fluid intake takes place on the job, and results in an estimated exposure dose of 1.4 rem and 0.11 rem to the skeleton and total body, respectively. The Tennessee River is used as a municipal water supply and, consequently, the 14-year old is the likely critical population group; the estimated dose from drinking this water is shown in Table 10. Dose from recreational use of the environment (listed in Table 29) is based on the following assumptions: an exposure time of 100 hours per year; an attenuation of bottom sediment radiation by three feet of water; the use of average concentrations of radionculides found in water and sediments to estimate dose for periods when data are lacking; and the adsorption of beta particles by the flesh of man thus limiting the exposure of the skeleton to gamma radiation. Only the feet of the swimmer could be totally exposed to the radiation from contaminated bottom sediments, but this would not exceed about 30 times the dose from recreational exposure given in Table 29. Occupational exposure from work within a water treatment plant is not significantly different from background radiation (see Table 22) and, therefore, is not considered in total exposure dose estimation.

The estimated dose from intake of contaminated fish, and the fraction of MPI attained by standard man from consuming 37 lbs per year of the flesh of contaminated bottom feeders is about equal to that from drinking contaminated water. However, the average fish consumption in the South is

TABLE 29

ESTIMATED CUMULATIVE DOSE RECEIVED BY CRITICAL ORGANS OF MALES FROM USE OF CLINCH RIVER AND TENNESSEE RIVER[a]

(rem)

Critical	Clinch River		Tennessee River	
Pathway	Skeleton	Total Body	Skeleton	Total Body
Drinking Water	1.4	0.11	0.38	0.030
Recreation	0.018	0.019	0.003	0.003
Fish	1.8	0.14	0.070	0.0057
Total	3.2	0.27	0.45	0.039
Maximum Permissible Dose[b]	60	10	20	1.0

[a]Aggregate exposure for the period 1944 to 1963.

[b]As recommended by ICRP (see references 4 and 8), the annual dose rates for continuous occupational exposure are reduced to 1/10 and applied to the Clinch River and are reduced to 1/30 for bone as critical organ and to 1/100 for total body as critical organ and applied to the Tennessee River.

24 lbs per year.[58] As a likely approximation, it is assumed that the total dose from eating Clinch River fish is 24/37 of the dose due to drinking Clinch River water and amounts to 1.8 rem and 0.14 rem to the skeleton and total body, respectively. It is further assumed that bottom feeders taken from the Tennessee River are diluted with other East Tennessee fish, and result in a total dose of 0.070 rem to the skeleton and 0.0057 rem to the total body. Thus, the estimated total dose in the skeleton of the 18-year old utilizing the Clinch River is 3.2 rem. The estimated dose to the skeleton of the 14-year old residing along the Tennessee River is 0.45 rem. In both cases the dose estimate is less than one-tenth of the maximum permissible dose. These estimated doses are believed to be high as a result of the conservative assumptions made in their estimation.

POTENTIAL EXPOSURES FROM CROP IRRIGATION

Irrigation of a variety of crops has been practiced in Tennessee for at least 50 years.[37] A survey conducted in 1958 by the Tennessee Division of Water Resources indicated that 1021 irrigation units were in operation at that time on about 0.5% of the crop lands; 20% were used for irrigating truck crops and 30% for irrigating feed crops (corn, silage, and hay). Ground water is the principal source of irrigation water west of the Tennessee River, and surface water is predominantly used in East Tennessee. More than 15 inches of water has been applied to truck crops during the growing season.

At present there is no crop irrigation along the Clinch River.[38] Therefore, an analysis of the possible consequences of transfer of fission products from contaminated river water to foods by crop irrigation is an hypothetical exercise. However, safety analyses are expected to point out future problem areas as well as assess the safety of current practice. Thus, the justification for such an exercise lies in uncovering any long-term problems that may be associated with usage of this natural resource.

Direct measurements of soil or crop loading with fission products due to irrigation practice, as distinguished from fallout and rainout are not available. It is necessary, therefore, to estimate the exposure dose that man may receive from intakes by this path on the basis of assumptions on soil loading, transfer coefficients from soil to crop, foliar contamination, and dietary habits of man.

Soil Load

Ion Exchange Reactions and Parameters

When water containing fission products is passed through a soil, the radioactive cations are removed from solution by ion exchange and their movement is restricted. Stable ions also exchange with those previously on the soil.

In the case of a divalent-monovalent ion system, it was assumed that all divalent ions behave similarly. The monovalent ions compete with the divalent ions for the exchange sites according to the expression, $2MR + D^{++} = DR_2 + 2M^+$, where D and M stand for divalent and monovalent notation.[39] The selectivity coefficient of divalent to monovalent ions is represented by[40]

$$K_M^D \text{ (g/ml)} = \left(\frac{q_D}{C_D}\right)\left(\frac{C_M}{q_M}\right)^2 \tag{20}$$

where

K_M^D is the selectivity coefficient of D with respect to M, q_D and q_M are the partial exchange capacities for D and M(meq/g) and C_D and C_M are the equilibrium concentration of D and M in solution (meq/ml).

From equation 20 and taking the total exchange capacity $Q = 0.15 \frac{\text{meq}}{\text{g}}$ $(Q = q_M + q_D)$, the selectivity coefficient $K_M^D = 30$ g/ml,[41] and the concentration of stable ions in Clinch River and Tennessee River water $C_D = .00164 \frac{\text{meq}}{\text{ml}}$ (calcium plus magnesium) and $C_M = .000142 \frac{\text{Meq}}{\text{ml}}$ (sodium plus potassium), the partial divalent cation capacity of the soil is calculated to be $0.1497 \frac{\text{meq}}{\text{g}}$. Even if the selectivity coefficient were as low as unity, the divalent cations would occupy over 99% of the exchange sites because of valence effects.

The selectivity coefficient of strontium to calcium on the exchanger can be expressed by

$$K_{Ca}^{Sr} = \left(\frac{q_{Sr}}{C_{Sr}}\right) \quad \left(\frac{C_D}{q_D}\right) = K_{d_{Sr}} \left(\frac{C_D}{q_D}\right) \tag{21}$$

where

$K_{d_{Sr}}$ = (distribution coefficient) is the ratio of the concentration of strontium sorbed per unit weight of exchanger to the concentration of unsorbed strontium per unit volume of solution, at equilibrium

By the use of the distribution coefficient, an estimate can be made of the soil loading of a particular radionuclide at equilibrium. From equation 21, assuming the strontium will not affect the partial exchange capacity $q_D = 0.1497$, and taking $K_{Ca}^{Sr} = 1.3$,[40] and the concentration of stable calcium plus magnesium $C_D = 0.00164\ \frac{meq}{ml}$, the distribution coefficient for strontium is calculated to be 120 ml/g. From measurements, an average $K_{d_{Sr}}$ of 110 $\frac{ml}{g}$ is reported for Clinch River sediments.[43]

By similar considerations, the selectivity coefficients of cesium to sodium on the exchanger can be expressed as

$$K_{Na}^{Cs} = \left(\frac{q_{Cs}}{C_{Cs}}\right) \left(\frac{C_M}{q_M}\right) = K_{d_{Cs}} \left(\frac{C_M}{q_M}\right) \qquad (22)$$

where

$K_{d_{Cs}}$ = (distribution coefficient) the ratio of the concentration of cesium sorbed per unit weight of exchanger to the concentration of unsorbed cesium per unit volume of solution, at equilibrium.

For the case of cesium exchange by local Conasauga shale, the estimation of soil loading is more involved. There are small quantities of exchange sites (fixation sites) highly selective for the heavy alkali metal cations (K^+ through Cs^+), and the selectivity for cesium compared to the stable ions of the system varies with the relative concentrations. Consequently, the use of equation 20 to estimate the partial monovalent loading would be misleading, since it would indicate a low value for cesium loading when substituted in equation 22.

Study of cesium exchange by Conasauga shale indicates that the number of exchange sites highly selective for cesium amounts to about 0.013 $\frac{meq}{g}$.[44] For the total exchange complex it is found that for $\frac{C_{Cs}}{C_{Na}} = 10^{-5}$ (typical of Clinch and Tennessee River water), $K_{Na}^{Cs} = 2000$. Because the majority of the exchange sites have little affinity for cesium compared to sodium, practically all of the cesium would be held at the fixation sites; the cesium to sodium selectivity coefficient of the fixation sites is estimated to be approximately $\frac{0.15}{0.013}$ x 2000 or 2.3×10^4. No valence effect is observed for the fixation sites; however, potassium is found to be approximately 10 times and calcium 9 times as effective as sodium in inhibiting the sorption of cesium. Magnesium is assumed to be as effective as calcium in restricting cesium sorption. The effective concentration of competing cations is taken as $C_M = C_{Na} + 10\ C_K + 9\ C_{Ca} + 9\ C_{Mg} = 0.015\ \frac{meq}{ml}$. Since $q_{Cs} >> q_M$, q_M is assumed to be equal to the concentration of fixation sites.

From equation 22, when $K_{Na}^{Cs} = 2.3 \times 10^4$, $q_M = 0.013\ \frac{meq}{g}$, and $C_M = 0.015\ \frac{meq}{ml}$, the distribution coefficient is calculated to be $2.1 \times 10^4\ \frac{ml}{g}$. An average $K_{d_{Cs}}$ for Clinch River sediments is reported to be $2.9 \times 10^4\ \frac{ml}{g}$.[32]

Estimation of Soil Loading

The fission product loading of the soil is estimated by assuming that the soil will continue to remove all applied exchangeable cations until saturated to the equilibrium value. The volume of irrigation water required to attain equilibrium may be calculated by use of the distribution coefficients and the mass of soil available. Assuming a soil depth of 6 2/3 inches and a soil density (dry weight) of 1.32 g/cm^3, the estimated soil mass per square meter is 2.24×10^5g. The depth of irrigation water required to attain equilibrium for ^{90}Sr is given by:

$$120\ \frac{ml}{g} \times 2.24 \times 10^5\ \frac{g}{2} \times 3.28\ \frac{ft}{m} \times 10^{-6}\ \frac{m^3}{cm^3} = 88\ ft$$

Similarly, for ^{137}Cs, 16,000 feet of water would be required to reach equilibrium. Water applied after the soil reaches its equilibrium load is assumed to have little additional effect on soil loading.

In an operating irrigation system, the accumulation of any particular fission product in the soil over a differential time element, dt, is assumed to be expressed by the equation:

$$\frac{dN(t)}{dt} = R - \lambda N(t) - aN(t) - \beta N(t) \qquad (23)$$

where

$N(t)$ = quantity of radionuclide $\left(\frac{\mu c}{m^2}\right)$,

R = rate of application of the fission product $\left(\frac{\mu c}{yr\text{-}m^2}\right)$,

λ = decay constant (yr^{-1}) of the fission product,

a = fractional loss per year of fission product to the crop (yr^{-1}),

β = fractional loss per year of fission product due to other causes such as soil erosion, leaching, etc (yr^{-1}).

The loss of ^{90}Sr and ^{137}Cs from the soil zone by erosion, leaching, surface runoff, and interflow, is assumed to be negligible. In reality, some loss by these machanisms is expected. Numerous accounts of the occurrence of ^{90}Sr and ^{137}Cs in soils indicate only a slow movement of these radionuclides through the soil by leaching.[45-50] Loss of ^{90}Sr in fallout due to erosion and runoff is related to the soil cover and the land slope on plots growing agricultural crops. Investigation of plots in Wisconsin and Georgia indicate losses by erosion and runoff ranging from 0.4% to 4.0%.[51] Studies are currently in progress at ORNL to define the loss of radionuclides by erosion and runoff from local soil plots. The solution of equation 23 when

$N(t) = 0$ at $t = 0$, and $\beta = 0$ is:

$$N(t) = \frac{R}{\lambda + a} \left(1 - e^{-(\lambda + a)\,t}\right) \qquad (24)$$

The rate of application of fission products to the agricultural plot is a function of the quantity of irrigation water used and the concentration of fission products in the water. Tables 30 and 31 list the build-up of ^{90}Sr and ^{137}Cs in soil resulting from an assumed rate of application of 2 ft of Clinch River water and Tennessee River water per year. The concentration of ^{90}Sr in the water is assumed to be constant, either at the level that occurred in 1951 or at the level that occurred in 1954. Similarly, ^{137}Cs concentrations are assumed constant at the level of 1953 or at the level of 1956. Separate land areas are assumed to support the growth of grain, leafy vegetables, potatoes, or pasture grass. Values chosen for the fractional loss (a) of ^{90}Sr and ^{137}Cs to the various crops are discussed below. Due to radioactive decay and to the fractional loss to the crop, ^{137}Cs essentially reaches equilibrium in the soil system after 100 years of irrigation. An external dose rate of less than 0.1 mrad/day is associated with the contaminated soil at equilibrium.

Transfer of Radionuclides to Man through Soil

Plant growth requires that ions from the soil be removed continuously and relocated within the plant. This dynamic system allows fission products in the soil to be transferred to the plant. Values reported for ^{90}Sr and ^{137}Cs transferred from soil to crop vary by a factor of about 10.[52-57] Many of the transfer coefficients result from experimental studies that require an extrapolation to field conditions. Selection of transfer coefficients for this study consider the exchangeable calcium, the cation exchange capacity, the pH, and the exchangeable hydrogen of local soils. An estimated plant load ($\frac{\mu c}{kg}$ dry weight) is based on soil to crop transfer of 0.01% of the ^{137}Cs (all crops), 0.005% of the ^{90}Sr to wheat grain, and 1% of the ^{90}Sr to other crops, and an edible crop yield of 0.14 kg/m^2 dry weight for wheat grain (typical for East Tennessee) and one kg/m^2 for all other crops.

Estimated Intake of ^{90}Sr and ^{137}Cs

The daily intake of ^{90}Sr and ^{137}Cs is estimated by assuming that all produce for the year comes from the same irrigated soil, and that the dietary habits of the individual include an average daily intake of 0.24 kg of grain, 0.26 kg of leafy vegetable, and 0.1 kg of potatoes.[58] Most of the wheat grain is in the form of white flour; therefore, only 25% of the ^{90}Sr in grain is expected to reach the flour and be consumed by man.[59]

By assuming that the maximum permissible intake (MPI) of a radionuclide ($\frac{\mu c}{day}$) is simply the product of the $(MPC)_w$ and the volume of water consumed by the standard man (2200 ml/day), the fraction of MPI that may be attained by consuming contaminated produce is calculated (Table 32 and 33). Inherent in the calculation is the assumption that rainfall will not affect the soil loading and that the fission products will be uniformly distributed within the soil by land cultivation procedures.

TABLE 30

THE ESTIMATED CUMULATION OF ^{90}SR IN IRRIGATED SOIL
(10^{-2} $\mu c/m^2$)

Years of Irrigation	1951 Type of Crop: Grain	1951 Type of Crop: Leafy Vegetable or Grass	1951 Type of Crop: Potato	1954 Type of Crop: Grain	1954 Type of Crop: Leafy Vegetable or Grass	1954 Type of Crop: Potato
CLINCH RIVER MI 14.5						
1	0.30	0.30	0.31	2.9	2.9	3.0
2	0.60	0.58	0.62	4.9	5.7	6.1
5	1.4	1.3	1.5	14	13	14
11	2.8	2.3	2.9	28	22	28
30	4.7	3.4	5.9	56	34	57
44	6.9	3.6	7.1	67	36	69
TENNESSEE RIVER MI 465.5						
1	0.050	0.050	0.052	0.33	0.33	0.35
2	0.10	0.098	0.11	0.67	0.65	0.70
5	0.24	0.22	0.24	1.6	1.4	1.6
11	0.48	0.38	0.49	3.2	2.5	3.2
30	0.96	0.58	0.99	6.4	3.9	6.6
44	1.2	0.61	1.2	7.7	4.1	7.9

TABLE 31

ESTIMATED CUMULATION OF ^{137}CS IN IRRIGATED SOIL

(10^{-2} $\mu c/m^2$)

Years of Irrigation	1953 Type of Crop: Grain	1953 Type of Crop: Leafy Vegetables, Potatoes, or Grass	1956 Type of Crop: Grain	1956 Type of Crop: Leafy Vegetables, Potatoes, or Grass
CLINCH RIVER MI 14.5				
1	0.096	0.095	2.2	2.2
5	0.42	0.37	9.8	8.7
10	0.71	0.57	17	13
30	1.2	0.79	28	18
50	1.3	0.81	31	19
100	1.4	0.81	32	19
TENNESSEE RIVER MI 465.5				
1	0.015	0.015	0.36	0.36
5	0.065	0.058	1.6	1.4
10	0.11	0.090	2.7	2.2
30	0.19	0.12	4.6	3.0
50	0.21	0.13	5.0	3.1
100	0.21	0.13	5.2	3.1

TABLE 32

ESTIMATED FRACTION OF ^{90}SR MPI THAT MAN MAY ATTAIN BY TRANSFER FROM SOIL CONTAMINATED WITH IRRIGATION WATER (10^{-1} MPI)[a]

Years of Irrigation	1951 Grain	1951 Leafy Vegetables	1951 Potato	1954 Grain	1954 Leafy Vegetables	1954 Potato
CLINCH RIVER MI 14.5						
1	0.0007	0.086	0.035	0.0070	0.85	0.34
2	0.0015	0.017	0.072	0.014	1.7	0.70
5	0.0035	0.39	0.17	0.034	3.7	1.6
11	0.0069	0.65	0.33	0.067	6.4	3.2
30	0.014	1.0	0.67	0.14	9.8	6.6
44	0.017	1.1	0.81	0.16	10	8.0
TENNESSEE RIVER MI 465.5						
1	0.0004	0.049	0.020	0.0027	0.32	0.13
2	0.0008	0.095	0.040	0.0054	0.63	0.27
5	0.0020	0.21	0.094	0.013	1.4	0.62
11	0.0042	0.37	0.19	0.026	2.4	1.2
30	0.0078	0.56	0.38	0.052	3.7	2.5
44	0.0094	0.59	0.44	0.062	4.0	3.0

[a]Considering bone as the critical organ.

TABLE 33

ESTIMATED FRACTION OF ^{137}CS MPI THAT MAN MAY ATTAIN BY TRANSFER FROM SOIL CONTAMINATED WITH IRRIGATION WATER (10^{-4} MPI)[a]

Years of Irrigation	1953 Grain	1953 Leafy Vegetables or Potato	1956 Grain	1956 Leafy Vegetables Or Potato
CLINCH RIVER MI 14.5				
1	0.0013	0.0077	0.031	0.18
5	0.0051	0.030	0.13	0.71
10	0.0096	0.047	0.23	1.1
30	0.017	0.064	0.39	1.4
50	0.018	0.066	0.42	1.5
100	0.019	0.066	0.44	1.5
TENNESSEE RIVER MI 465.5				
1	0.0020	0.012	0.049	0.29
5	0.0088	0.047	0.22	1.2
10	0.015	0.073	0.37	1.8
30	0.026	0.099	0.63	2.4
50	0.028	0.10	0.69	2.5
100	0.029	0.10	0.70	2.5

[a]Considering total body as the critical organ.

After 44 years of irrigation with waters containing a constant concentration of ^{90}Sr an equilibrium is established and an increase in ^{90}Sr intake would not be expected. At the estimated rate of consumption of both grain and leafy crops, the hypothetical intake of ^{90}Sr at equilibrium using Clinch River water for irrigation may range from 0.19 to 1.8 times MPI; similarly, the hypothetical intake of ^{90}Sr at equilibrium using Tennessee River water may range from 0.10 to 0.71 times MPI. At the current levels of ^{137}Cs in Clinch River and Tennessee River water, no apparent problems will be encountered.

Experimental results from a number of studies of radionuclide uptake by plants are summarized by the Stanford Research Institute.[60] Transfer of radionuclides from soil to crop is expressed as a soil uptake contamination factor, A_{su} or the ratio of atoms per gram of dry plant to the atoms per gram of soil. By considering the characteristics of East Tennessee soils, average values of $A_{su}(^{90}Sr)$ are calculated for various crops as follows: grain, 0.05; potatoes, 0.26 dark green and deep yellow vegetables (spinach, broccoli, carrots, etc.), 3.3; other green vegetables (beans, peas, lettuce, etc.), 4.6; and other vegetables (beets, radishes, etc.), 3.6. The daily intake of ^{90}Sr is estimated from the dietary habits in the South and the soil loading at equilibrium (Table 30, Clinch River, 1954).[58] Based on the estimated daily intake of ^{90}Sr, a standard man is calculated to attain about two times the MPI due to soil to crop transfer. Another comparison of calculated values of ^{90}Sr content in edible foods is afforded by the Federal Radiation Council.[61] They predict an average accumulation of 0.044 μc of ^{90}Sr per square meter from fallout at the end of 1963, and 250 pc of ^{90}Sr per kilogram in harvested wheat. Using the assumptions of soil to grain transfer and productivity previously listed, the estimated ^{90}Sr in grain is 15 pc per kilogram. Thus about 5% of the ^{90}Sr expected in wheat grain may come from the soil and about 95% from absorption through above ground parts of the plant. Menzel reports similar values for foliar retention of ^{90}Sr by wheat grain.[62] The importance of contaminated irrigation water as a critical pathway receives support from experimental field studies reported by Michon.[63] Results indicate the average ^{90}Sr content in one kilogram of the crops studied is equivalent to the ^{90}Sr in at least 9.5 liters of the irrigation water.

Transfer of Radionuclides by Foliar Contamination

To a large extent crop irrigation is accomplished by spray techniques. Thus, foliar contamination of above ground crops is another avenue by which fission products in irrigation water may reach man. Studies of aerial contamination of plants have been related principally to absorption of fallout radionuclides; therefore, uncertainties exist in applying data from such studies to crop contamination by irrigation. The amount of radionuclide accumulated in edible parts of a plant depends on the stage of growth of the plant at the time of spraying, and the rate of translocation and rate of accumulation of the radionuclide.[64] Intermittant rainfall is known to remove a fraction of the radionuclide previously deposited on plants. The values of percent retention by the edible part of plants vary considerably between different crops. Grain is reported to contain from 0.2% to 2% of the ^{90}Sr, and apparently depends on the season and time of contamination.[62,65-67] The final content of ^{137}Cs in wheat grain ranges from 1% to 5%. Potatoes contain significantly smaller amounts of ^{90}Sr than other vegetable crops; the final content in tubers ranges from 0.01% to 0.04%. Cesium-137 retention by tubers averages about 9%.[68] Estimates of retention by leafy vegetables are based on values reported for pasture grass and the wheat plant. Retention of ^{90}Sr varies between 2.5% and 5%, and ^{137}Cs retention is about 10%.[68,69]

Values selected for foliar retention of ^{90}Sr and ^{137}Cs for this analysis are based on the above information and are listed in Table 34. By irrigating the various crops with two feet per year of Clinch River and Tennessee River water that contain concentrations of ^{90}Sr and ^{137}Cs previously listed, the fraction of MPI that might be attained due to foliar contamination is calculated (Table 34). Foliar contamination with ^{90}Sr has a greater influence on leafy crops than grain or potato crops. At the levels of ^{137}Cs encountered in the rivers during 1953 and 1956, a smaller but perceptable amount of ^{137}Cs may enter man's diet due to foliar contamination as contrasted to soil to plant contamination.

Transfer of Radionuclides to Man by Milk

One final possibility of radionuclide transfer to man by contaminated

TABLE 34

TRANSFER OF ^{90}SR AND ^{137}CS TO MAN BY FOLIAR CONTAMINATION FROM IRRIGATION WATER

Source	Year	Percent Retention[a]			Plant Load[b] (10^{-4} μc/kg)			Fraction of MPI[c]		
		Grain	Leafy Vegetables	Potato	Grain	Leafy Vegetables	Potato	Grain	Leafy Vegetables	Potato
STRONTIUM-90										
CRM 14.5	1951	1.0	5.0	0.03	2.2	1.6	0.012	0.015	0.045	0.00014
CRM 14.5	1954	1.0	5.0	0.03	22	15	0.092	0.15	0.45	0.0011
TRM 465.5	1951	1.0	5.0	0.03	.38	0.27	0.0016	0.0087	0.033	0.00006
TRM 465.5	1954	1.0	5.0	0.03	2.5	1.8	0.011	0.058	0.17	0.0004
CESIUM-137										
CRM 14.5	1953	5.0	10	10	3.6	1.0	1.0	0.0020	0.00059	0.00022
CRM 14.5	1956	5.0	10	10	84	23	23	0.046	0.014	0.005
TRM 465.5	1953	5.0	10	10	.56	0.16	0.16	0.0031	0.0009	0.0004
TRM 465.5	1956	5.0	10	10	14	3.8	3.8	0.074	0.022	0.009

[a]Contents of edible plant as % of radionuclide applied per unit of land area.

[b]kg of dry weight

[c]Assuming a daily intake of man of 0.24 kg of grain, 0.26 kg of leafy vegetables, and 0.10 kg of potatoes.

irrigation water is considered; that is, the transfer of the radionuclides into the milk of cows allowed to graze on pasture land irrigated with river water. Information assembled by the United Nations Scientific Committee on the Effects of Atomic Radiation indicates that 0.08% of the ^{90}Sr and 1.3% of the ^{137}Cs ingested daily by dairy cattle is transferred to each liter of milk.[70] The intake of these radionuclides by the cow is based on an average daily consumption of 10.9 kg of pasture grass.[71]

Using the previously calculated values of leafy crop loading due to soil to crop transfer and direct foliar contamination, and assuming that one liter per day of milk is consumed, the fraction of MPI that may be obtained by drinking milk is estimated (Table 35). The hypothetical intake of ^{90}Sr at equilibrium using Clinch River water and Tennessee River water for irrigation may result in an MPI ranging from 0.0052 to 0.051 MPI and 0.0029 to 0.019 MPI, respectively. At the levels of ^{137}Cs in the irrigation water, no apparent problem is encountered. Since foliar retention is the principal mechanism of ^{137}Cs transfer to grass, there is little change in loading from year to year. At equilibrium, the fraction of MPI (total body as critical organ) attained is: Clinch River, 3.2×10^{-4} (1953) and 7.6×10^{-3} (1956); and Tennessee River, 5.1×10^{-4} (1953) and 1.2×10^{-2} (1956).

Cumulative Transfer of Radionuclides to Man by Crop Irrigation

Although independent consideration was given to the several vectors of contaminating man's diet by irrigating water, these vectors are additive. Hypothetically, the cumulative contribution to the MPI (bone) of ^{90}Sr by irrigation water is shown in Figures 14 and 15. Similarly, the cumulative contribution to the MPI (total body) of ^{137}Cs by irrigation water is shown in Figure 16. Calculations indicate that soil to crop transfer of ^{90}Sr may be of greatest long-term importance in contributing ^{90}Sr to man's diet from contaminated irrigation water. Foliar and milk vectors may be only of secondary importance. After equilibrium is attained in the soil, standard man might ingest 20 times as much ^{90}Sr by consuming produce from the land irrigated with contaminated water compared to consumption of the water. No apparent problem is indicated due to the estimated concentration of ^{137}Cs in Clinch River and Tennessee River

TABLE 35

ESTIMATED FRACTION OF ^{90}SR MPI THAT MAN MAY ATTAIN FROM MILK AFFECTED BY CROP IRRIGATION[a]

Years of Irrigation	CLINCH RIVER Mi. 14.5		TENNESSEE RIVER Mi. 465.5	
	1951	1954	1951	1954
1	0.0019	0.018	0.0010	0.0069
2	0.0021	0.021	0.0012	0.0080
5	0.0028	0.028	0.0016	0.011
11	0.0038	0.037	0.0021	0.014
30	0.0050	0.049	0.0028	0.019
44	0.0052	0.051	0.0029	0.019

[a]Assuming a daily intake by man of one liter of milk and considering bone as the critical organ.

irrigation water. However, due almost entirely to foliar contamination, as much as 30 times the ^{137}Cs might be ingested from contaminated crops compared to drinking water. It is not possible to determine the accuracy of these predictions with the information currently available. A number of assumptions were necessary in making the calculations, and differences in the values used for transfer parameters could significantly change the estimated intake of ^{90}Sr and ^{137}Cs. For example, the loss of radionuclides from irrigated plots by erosion and runoff may reduce the quantity of radionuclides available to the plants. A change in soil to crop transfer, in foliar retention, or in productivity of edible crops, could either increase or decrease the quantity of ^{90}Sr and ^{137}Cs present in man's diet. Differences in dietary habits and use of produce from uncontaminated plots would also influence the estimated internal exposure.

At the present time no problem exists of significant quantities of fission products entering man's diet due to irrigation practice. It seems unlikely that a problem will develop along the Clinch-Tennessee River system. Truck crops in this environment contribute little to the total quantity of produce and are grown only for a short period during the year. However, crop irrigation with contaminated water could take on greater importance in areas where climatic conditions are more conducive to year-around irrigation of large agricultural plots. Some use can be made of data provided by fallout studies, but there is need for data from experiments designed to elucidate radionuclide entry into man's diet from contaminated irrigation water. In view of the long-range interest in a power reactor program and the accompanying recycle of fuels, such studies should consider the transmutation products (resulting from neutron capture by irradiated fuels), as well as fission products.

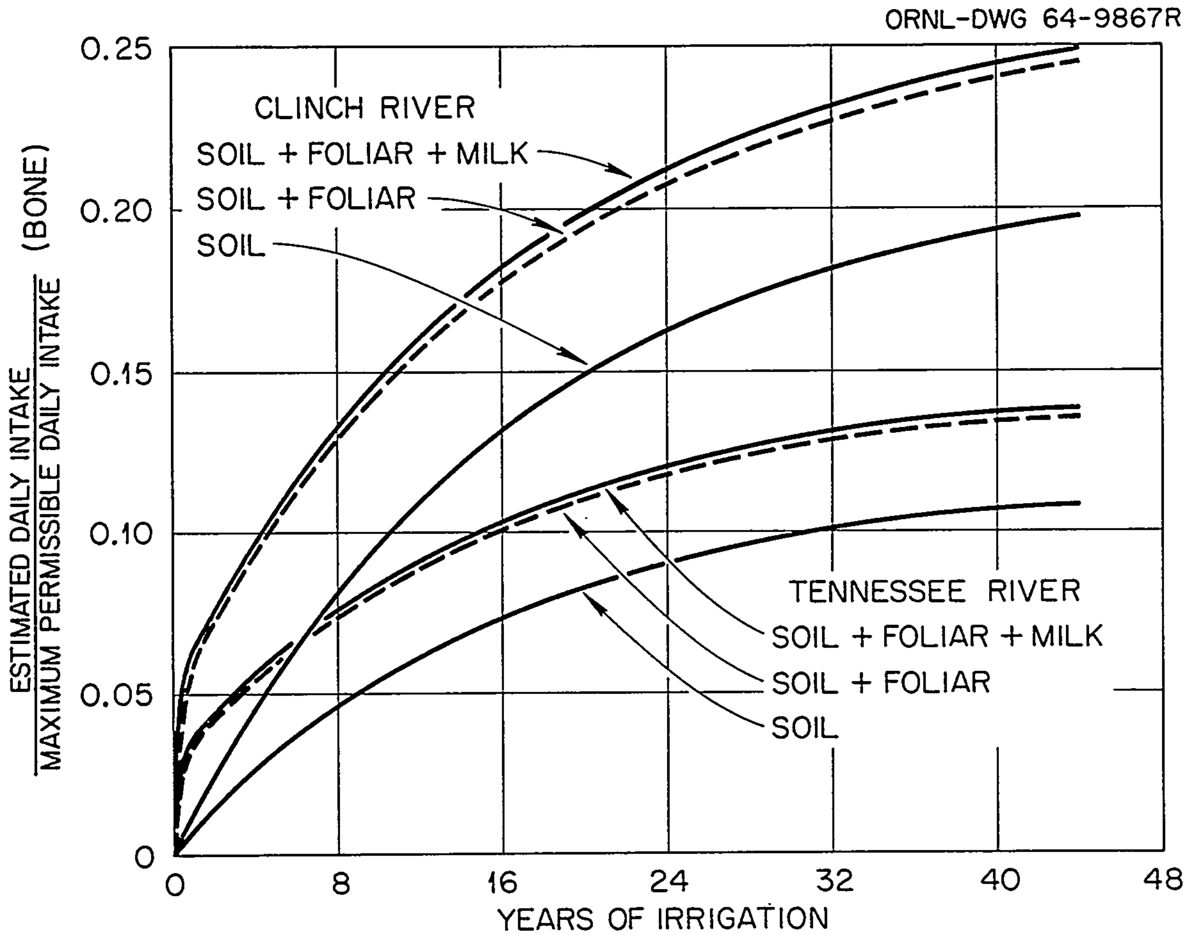


Fig. 14. Potential Contribution to MPI by Sr^{90} in Irrigation Water: 1951.

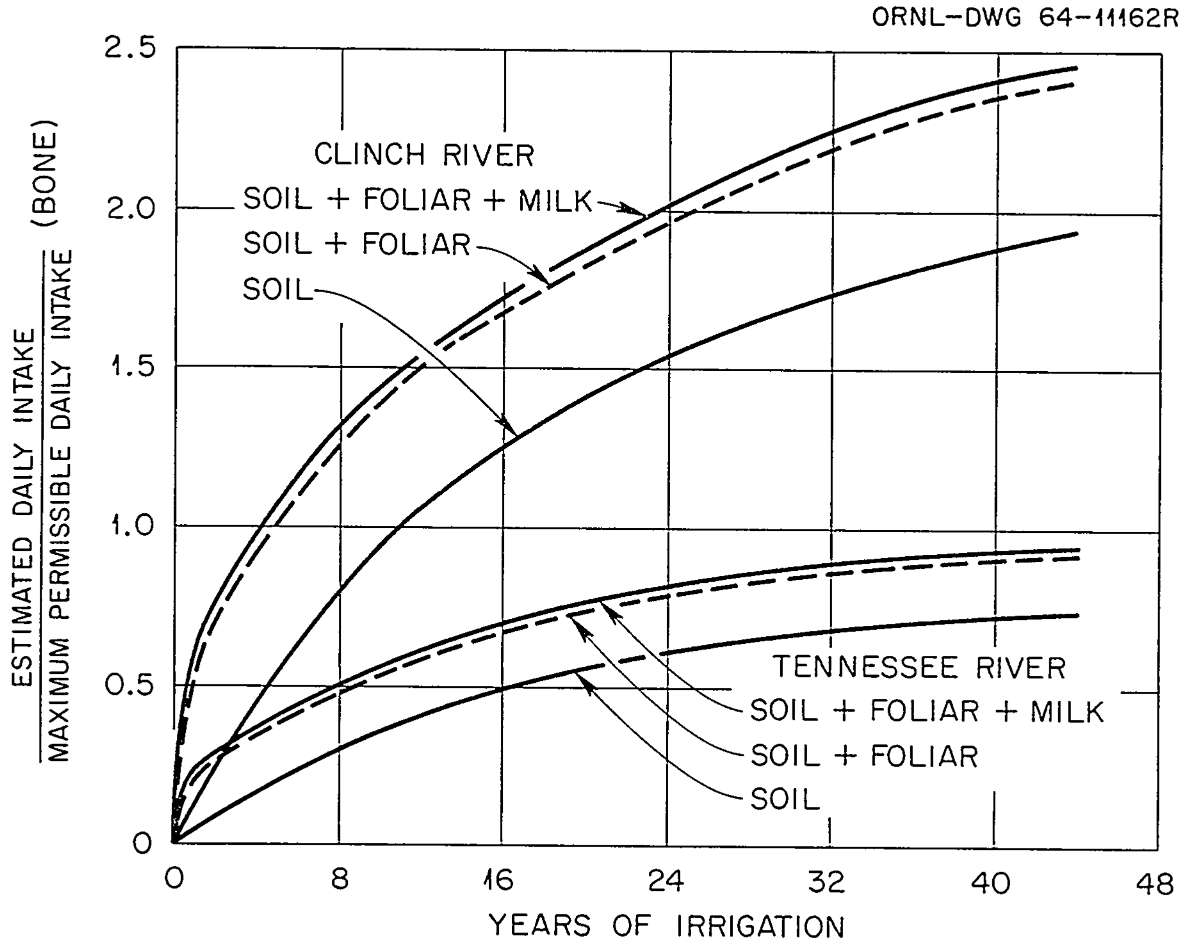


Fig. 15. Potential Contribution to MPI by Sr^{90} in Irrigation Water: 1954.

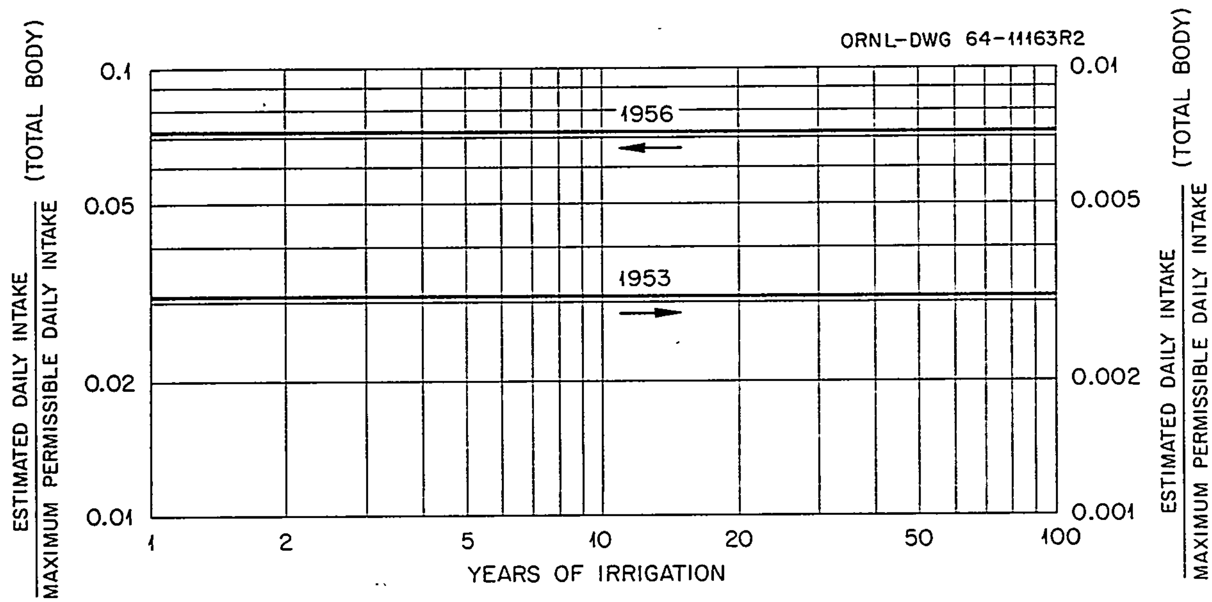


Fig. 16. Potential Contribution to MPI by Cs^{137} in Clinch River Irrigation Water.

CONCLUSIONS AND RECOMMENDATIONS

Disposal of radioactive wastes to the Clinch River has resulted in radiation exposures well below ICRP and FRC permissible limits. Of the critical pathways considered, external exposure from contaminated water and bottom sediments was of less importance as a potential source of radiation exposure than consumption of contaminated water and fish. However, if the practice of irrigation with river water develops, the long term effect of crop irrigation with contaminated water could become the most important avenue of exposure.

Internal dose estimations based solely on exposure of a standard man will underestimate the dose to critical population groups; that is, the most highly exposed group. By taking account of differences in rates of intake and masses of critical organs estimated doses exceed those of standard man by a factor of at least two. Such differences are in addition to those expected as a result of individual variability. Other improvements in dose estimates are possible but that will require better information on the habits and characteristics of population groups likely to be exposed. Such information includes the location and age distribution of potentially exposed populations, amounts of principal foodstuffs consumed, and their principal occupational and recreational habits. The type and rate of consumption of fish, potatoes, and leafy vegetables as a function of age would be extremely useful. In particular, additional information is necessary to decide if total fish (flesh and bone) are an important source of ^{90}Sr intake by man. Although the cooked flesh of fish is an established staple of man's diet, the consumption of total fish by East Tennessee fishermen is not confirmed.

Strontium-90 is the most important of the critical radionuclides in liquid wastes released to the Clinch River, contributing more than 99% of the skeleton and total body dose and 70% of the thyroid dose. Ruthenium-106, ^{137}Cs, and ^{60}Co contribute significantly to the dose received by the GI tract. As a consequence of ^{90}Sr releases, the skeleton of man drinking Clinch River water is the critical organ receiving about 5 times the total dose of the other organs considered. However, the total dose to the skeleton of the critical population group was considerably smaller (by a factor of about 20) than the allowable dose from contaminated drinking water. Improved waste management at ORNL has resulted in a decrease in ^{90}Sr released to the Clinch River. The more recent discharges to the river have been about equal to the contribution from nuclear test fallout.

An internal dose commitment is created for the future by the intake of radionuclides of long effective half life. Dose continues to be delivered to the critical organs following intake and depends on the effective half life of the radionuclide. Information on dose commitment may be useful if changes in population exposure limits are considered, if a new installation wishes to utilize the diluent capacity of a surface water, or if an accidental release of radioactive material requires corrective action. Methods developed in this report for estimating dose to man can be applied to the assessment of future radiation exposure.

Greatest emphasis of routine environmental monitoring should be related to current and critical pathways of exposure; for man, these are the consumption of water and fish. Periodic evaluation is needed to confirm the adequacy of the monitoring program and to reestablish the importance of critical nuclides and critical exposure pathways. Such review would be concerned not only with radionuclides of long physical half life, but also with those of short half life that may occasionally be released and otherwise overlooked by a routine program. The routine monitoring program should include comparison of gross beta analysis made on daily samples and on monthly composite samples, the difference in magnitude being indicative of the significance of the contribution of short lived radionuclides in the effluent. Although contaminated water and bottom sediments are a minor source of radiation exposure to man, direct measurements of radiation intensity are desirable initially to confirm dose-rate calculations and to occasionally reconfirm the source potential. It is also desirable to investigate current and possible future use of Clinch River and Tennessee River waters as sources of supplemental water for irrigation purposes. This information can be used to define the need for sampling soils or crops in the affected areas.

There is need for additional research on areas of uncertainty associated with radionuclide transfer to fish and to irrigated crops. Information such as the rate of transfer and quantity of ^{90}Sr and stable strontium in flesh and bone of important fish species, the influence of fish age and season of the year on transfer rates, and the transfer of ^{90}Sr from fish bone to fish flesh by cooking would be helpful to estimate the dose to man and to optimize a fish monitoring program. A potentially important source of ^{90}Sr entry in man's food chain can be eliminated by preventing fish in White Oak Creek or White Oak Lake from entering the Clinch River. Analysis of crop irrigation as a critical exposure pathway requires knowledge of fission product behavior in soils and plants. Of greatest importance is information applicable to the East Tennessee environs. This includes the accumulation of fission products in cultivated soils with time, the transfer of fission products from soil to plant, and the foliar retention of fission products by the plant.

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